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This document reports on 1987 research activities of the National Acid Precipitation Assessment Program (NAPAP). It outlines 1987 research highlights, future research activities, major deliverables, and the program's 1990 research objectives. Its seven substantive chapters cover NAPAP research in the areas of "Emissions and Controls," "Atmospheric Chemistry," "Atmospheric Modeling and Transport," "Atmospheric Deposition and Air Quality Monitoring," "Terrestrial Effects" (on Forests and Crops), "Aquatic Effects," and "Effects on Materials and Cultural Resources." The document includes introductory materials on the National Program's organizational structure, efforts at research coordination, peer and program review activities, and budget. A complete listing of the Program's 1987 publications is also included.

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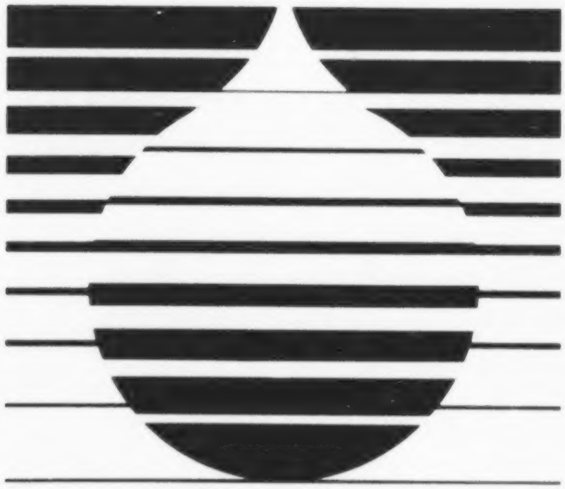

THE NATIONAL ACID PRECIPITATION ASSESSMENT PROGRAM

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National Acid Precipitation Assessment Program

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Environmental Protection Agency ● Department of Health and Human Services

Department of the Interior ● National Aeronautics and Space Administration

National Oceanic and Atmospheric Administration ● National Science Foundation

Department of State ● Tennessee Valley Authority

National Acid Precipitation Assessment Program
Office of the Director
722 Jackson Place, NW
Washington, DC 20503

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CONTENTS

EXECUTIVE SUMMARY

Emissions and Controls 1
Atmospheric Chemistry 2
Atmospheric Modeling and Transport 2
Atmospheric Deposition and Air Quality Monitoring 3
Terrestrial Effects 3
Aquatic Effects 3
Effects on Materials and Cultural Resources 4

INTRODUCTION

Background 5
NAPAP Assessment 6
Research Coordination 6
NAPAP Peer Review Activities 6
NAPAP Budget 9

RESEARCH PROGRESS AND OUTLOOK

Emissions and Controls 11
Atmospheric Chemistry 15
Atmospheric Modeling and Transport 23
Atmospheric Deposition and Air Quality Monitoring 29
Terrestrial Effects:
  Effects on Forests 35
  Effects on Agricultural Crops 38
Aquatic Effects 43
Effects on Materials and Cultural Resources 55

NAPAP PUBLICATIONS 63

EXECUTIVE SUMMARY

This report describes the research accomplishments of the National Acid Precipitation Program (NAPAP) during fiscal year 1987. The report supplements the NAPAP Interim Assessment as a summary of recent developments in the state of science as it pertains to the formation, transport, deposition, and effects of acidic and oxidant substances in the atmosphere. The research and monitoring activities of the National Program have been designed and conducted to rapidly advance the scientific basis for assessment activities and to provide the rational foundations for environmental policy as it relates to acidic deposition. As the Interim Assessment and five previous Annual Reports illustrate, the program has already produced an extensive body of research findings. Over the course of the next two years, the National Program will work with states, the private sector, and the domestic and international scientific communities to elaborate its research findings in an integrated assessment of the causes and effects of acidic deposition.

The past year has been especially active for the National Program because of the completion of the Interim Assessment and the conduct of ongoing research efforts. Work on the Assessment accelerated the examination of results of programs completed thus far. The Interim Assessment provides a foundation for organizing future research, and a vehicle to focus activity on those areas where a lack of understanding presently limits the credibility of predictions and extrapolations. The following list of program highlights illustrates advances made during the sixth year of research under the National Acid Precipitation Assessment Program.

EMISSIONS AND CONTROLS

To meet the needs of different users, the Emissions and Controls Task Group is continuing to produce both yearly current emissions trends data and a highly detailed inventory for a single year. Most of the effort has been directed toward the completion of a 1985 emissions data base and inventory. The emissions data base contains the estimated annual emissions from over 11,000 point sources and the inferred area source emissions for the more than 3000 counties in the contiguous 48 states. Starting with this annual emissions data base, the emissions inventory imposes a fine degree of temporal, chemical, and spatial resolution (typical hourly emissions of over 50 species for weekdays and weekends for each season over 20 km grid cells). This inventory serves as input to atmospheric models, such as the Regional Acid Deposition Model (RADM), which estimate how changes in emissions result in changes in deposition at downwind receptors. The Task Group continued to work with the Atmospheric Chemistry Task Group to integrate natural emissions data into this detailed emissions data base.

The Task Group continued its annual development of the yearly current trends inventory which has a state-level spatial resolution and a monthly temporal resolution for three species (SO_2, NO_x and VOC). The coverage of this inventory extends through 1986.

The Task Group continued to develop an integrated set of emissions projection models which will enable policy analysts to examine the emissions and costs of various control policies. Costs for retrofit reduction of SO_2 and NO_x emissions at 12 coal-fired power plants were developed and included in a draft report.

ATMOSPHERIC CHEMISTRY

This Task Group has continued its progress in all five of its research areas with results contributing to the work of other Task Groups, as well as providing information of direct relevance to decisionmakers. The Task Group reexamined natural emissions of sulfur from land sources in the eastern United States; natural emissions of sulfur from terrestrial sources in this region are now estimated to be only about 0.1 percent of anthropogenic emissions. Natural emissions of volatile organic compounds (VOC) were measured at one rural location in Pennsylvania. Model analysis of the results suggests that natural emissions of isoprene (a natural VOC) in that time and place contributed substantially to the production of rural ozone.

Comparisons of wet and dry sulfur and nitrogen deposition over several years are now available for a few research stations. At Argonne, Illinois, wet deposition of sulfur exceeded dry deposition in the autumn of two years, while wet and dry deposition were about equal in spring and summer. The winter patterns differed greatly, due primarily to a seven-fold variation in winter wet sulfate deposition. This variability was also observed in the winter for dry and wet nitrate deposition. At other times of the year, dry nitrate deposition was roughly equal to wet.

Research on Mountain Cloud chemistry and dry deposition illustrates patterns of deposition which are important with respect to high-elevation forests, and, in conjunction with other Task Group work, is serving to clarify the role of hydrogen peroxide (H_2O_2). Hydrogen peroxide is important because it may affect tree growth and because it is responsible for much of the aqueous-phase oxidation of SO_2. In winter the in-cloud concentrations of H_2O_2 are much lower than in the summer and less SO_2 is oxidized in winter than in summer. The winter hydrogen peroxide levels in the Northeast are low enough to support the notion of nonlinearity in the source-receptor relationship for sulfur. In the summer of 1986, hydrogen peroxide in the gas phase was 30 percent more abundant at a Virginia mountaintop site than at a similar site in New York. This observation is consistent with the hypothesis that higher light intensity and water vapor levels, such as are found further south, promote H_2O_2 formation. Task Group work also suggests that the total deposition of sulfate and nitrate increases with elevation, primarily as a result of increases in fog and cloud droplet interception.

ATMOSPHERIC MODELING AND TRANSPORT

The Atmospheric Modeling and Transport Task Group completed a major phase in the development of RADM (the Regional Acid Deposition Model). This "state-of-the-art" model will allow estimation of how changes in emissions result in changes in deposition at downwind receptors, taking into account the effects of nonlinearity. Preliminary evaluations have been completed and the Task Group continued to refine its plans for a two-year field program, the results of which will be used to evaluate the performance of RADM.

Current data indicate that on an annual basis, approximately 25 to 35 percent of the sulfur oxides and 15 to 25 percent of the nitrogen oxides emitted to the eastern North American atmosphere are transported out from the continent over the Atlantic Ocean. Research flights over the western Atlantic also found that ozone concentrations in the free troposphere (at distances of 200 to 300 km off the coast) are 30 to 40 ppb. The exception to this occurs near the base of clouds associated with a strong frontal system where concentrations are as high as 70 ppb.

Wet deposition in the area of the ASARCO smelter, a major SO_2 emitter near Seattle, Washington, was studied both when the smelter was operating and when it was shut down. The results show that the smelter accounts for about half of the wet sulfate deposition in the 600 km^2 area downwind of the smelter. However, the smelter's large contribution to the local wet deposition represents only a fraction, approximately 2 percent, of its total sulfur emissions. A further fraction of its sulfur emissions are deposited locally by dry deposition processes, while the remainder are transported and deposited in small amounts over much larger distances.

ATMOSPHERIC DEPOSITION AND AIR QUALITY MONITORING

The Atmospheric Deposition and Air Quality Monitoring Task Group continued to publish its summaries of wet deposition in the United States. The Task Group's analysis of the 150 station National Trends Network (NTN) showed that the network is capable of detecting statistically significant annual changes of 20 to 30 percent in hydrogen ion, sulfate, or nitrate concentrations in the eastern United States in regions with dimensions of 4 degrees of longitude and latitude (approximately 160,000 km^2). The sensitivity of the network varies from place to place. The first twenty stations in the rural air quality monitoring and dry deposition network were deployed this year. While dry deposition is more difficult to monitor than wet deposition, this network will provide an improved description of the patterns of total (wet and dry) acidic deposition.

Data from a site near Clingman's Dome, North Carolina, were compared to data from the MAP3S site at Oak Ridge, Tennessee. Results from this comparison suggest that the concentrations of major ions in wet deposition (e.g., H^+, $SO_4^=$, NO_3^-) decrease with elevation in the Great Smoky Mountains area, although precipitation amounts are nearly twice as high. Work at sites in the Mountain Cloud Chemistry Program, however, suggests that total deposition increases with altitude, reflecting increased fog and cloud droplet interaction with vegetation.

The chemical analysis of rain samples taken at remote stations around the world suggests that the average acidity of precipitation at these stations ranges from a pH of 4.7 to 5.2. Because the GTN measures organic acids while the NTN does not, the GTN and NTN results are not directly comparable for pH. Comparison of the GTN results with NTN data from the eastern United States does indicate that concentrations of ammonia, nitrate, and sulfate in wet deposition in the eastern United States are considerably higher than at the remote stations around the world.

TERRESTRIAL EFFECTS

The Terrestrial Effects Task Group continued to investigate the effects of acidic deposition and associated oxidants on forests throughout the United States. Studies indicate that natural stand aging is playing a major role in the observed declines of red spruce and balsam fir in low-elevation forests in New England. Other studies continue to characterize the essential association between roots and their fungal symbionts, and to determine whether this association may be altered by acidic deposition. At the current nitrogen deposition rate in the southeastern United States, nitrogen is not directly toxic to mycorrhizae.

Recent activities in crop research support the conclusion that ambient levels of precipitation acidity have little or no significant direct effects on crop yield under typical growing conditions. Ambient concentrations of ozone, on the other hand, have been demonstrated to have a damaging effect on crop yield.

For crops and forests, investigations continue to examine the interactions between acidic deposition and ozone, drought, insect infestations, and disease organisms. These complex interactions may have indirect and subtle effects on plant vitality.

AQUATIC EFFECTS

The Aquatic Effects Task Group continued to collect and analyse data from the National Surface Water Survey. Most acidic lakes in the Northeast are currently acidic because of mineral rather than organic acidity. However, it is not clear to what extent some of the lakes may have originally been acidic because of higher organic anion and lower base cation content. In another analysis of Eastern Lake Survey Data, subregional median lake sulfate concentrations were found to be highly correlated with estimated wet sulfate deposition in all but the Southern Blue Ridge Province. Results also show that there was no correlation between lake sulfate and the acid neutralizing capacity of lakes in the Northeast. This is probably because some watersheds compensate for acid deposition by releasing base cations, and thus ANC does not change in response to acidic deposition. Other evidence suggests, however, that in some lake systems, the acidic input may be only partially offset by release of base cations, in which case acidic deposition may reduce ANC.

Episodic acidification of lakes and streams continues to be a major focus of NAPAP research.

The relative importance of natural and anthropogenic factors in episodic acidification is being investigated. Models, based on existing or future surveys of chronic surface acidity, are being developed to predict the extent of acidic surface waters during episodes. A key component of the episodic studies is the evaluation of biological effects, with a focus on fish.

The Task Group continues to examine the watershed and in-lake factors controlling the response of surface waters to acidic input. These results complement the analyses being conducted in the Direct-Delayed Response Project, which will forecast changes in acid neutralizing capacity given certain levels of acidic deposition.

Field and laboratory research activities are conducted by the Task Group to better quantify the responses of fishes, invertebrates, birds, and other organisms to chemical changes associated with acidic deposition. The responses studied include mortality, bioaccumulation of metals, changes in community structure, and others. At the present time, atlantic salmon resources in the northeastern United States appear to be unaffected by acidic deposition.

Finally, the program is conducting continuing studies to determine the possible side effects of mitigative liming, as well as the role of acidic deposition in affecting drinking water quality in cisterns and shallow aquifers.

EFFECTS ON MATERIALS AND CULTURAL RESOURCES

In 1987 this Task Group continued to concentrate on the development of dose-response functions to relate changes in materials degradation rates to increments of acidic deposition and associated pollutants. The materials under examination are carbonate stone (marble and limestone), ten metals (including galvanized steel and bronze), and a variety of paint/substrate systems.

The Task Group is continuing its studies in laboratory chambers and at field exposure sites. At one field site, Research Triangle Park, North Carolina, preliminary calculations suggest that at ambient levels of deposition the dissolution of the zinc corrosion layer on galvanized steel due to the sum of dry deposition of SO_2 and wet deposition of sulfuric and nitric acids increases the dissolution caused by pure water in equilibrium with atmospheric CO_2 by less than 50 percent. The Task Group is also continuing work on the degradation of materials in place, and is planning the expansion of this work. The current focus of this activity is on cultural resources such as statues and monuments at Gettysburg National Military Park and an historic marble building in Philadelphia.

INTRODUCTION

BACKGROUND

This Annual Report to the President and Congress, submitted in accordance with the Acid Precipitation Act of 1980 (P.L. 96-294), describes the activities of the sixth full year of an integrated research effort of the Interagency Task Force on Acid Precipitation. The Task Force conducts the National Acid Precipitation Assessment Program which coordinates federally funded research and assessment activities to facilitate the timely development of a firm scientific basis for policy decisions that pertain to the phenomenon commonly referred to as "acid rain."

The Task Force is jointly chaired by the National Oceanic and Atmospheric Administration (NOAA); the Environmental Protection Agency (EPA); the Departments of Agriculture (USDA), Energy (DOE), and the Interior (DOI); and the Council on Environmental Quality (CEQ). The other participating Federal agencies are the Department of Health and Human Services (HHS), the Department of Commerce (DOC), the State Department (DOS), the National Aeronautics and Space Administration (NASA), the National Science Foundation (NSF), and the Tennessee Valley Authority (TVA). Membership in the Task Force also includes four Presidential appointees and representatives of Argonne, Brookhaven, Oak Ridge, and Pacific Northwest National Laboratories.

In addition to the Joint Chairs Council, the program's management structure is composed of a Director of Research, Task Group Leaders, and two Interagency committees. The Director of Research is charged with the executive management of the Program's research and assessment activities. The Task Group Leaders, who report both to their agencies and to the Director of Research, are responsible for the coordination of research in their particular subject areas. The two committees, the Interagency Science Committee (ISC) and Interagency Policy Committee (IPC), are made up of the Director of Research (an *ex officio* member of the IPC) and senior representatives of the agencies that make up the Joint Chairs Council (along with the Tennessee Valley Authority). The ISC and IPC are responsible for overseeing, respectively, the scientific quality and the policy relevance of the National Program's research and assessment activities.

Research activities under the National Program are carried out under seven Task Groups. The areas addressed by the Task Groups are: Emissions and Controls, Atmospheric Chemistry, Atmospheric Modeling and Transport, Atmospheric Deposition and Air Quality Monitoring, Terrestrial Effects on Forests and Crops, Aquatic Effects, and Effects on Materials and Cultural Resources.

The individual chapters within this report delineate each Task Group's primary research objectives, its 1987 research activities, highlights from 1987 research results, a discussion of future research activities and major deliverables, and Task Group-funded publications in 1987.

NAPAP ASSESSMENT ACTIVITIES

In September 1987, NAPAP released its Interim Assessment on the Causes and Effects of Acidic Precipitation. The four-volume document, which was prepared by 26 authors and reviewed by 68 external experts, is a comprehensive assessment of the "state of the science" of acidic deposition. While drawing mainly upon NAPAP research, the document also takes into account published information on relevant research conducted elsewhere in the United States and abroad. Its chapters cover the topical areas of "Emissions of Acidic Deposition Precursors," "Emission Control Technologies," "Future Emissions," "Atmospheric Processes," "Acidic Deposition and its Gaseous Precursors," "Effects on Agricultural Crops," "Effects on Forests," "Effects on Aquatic Systems," "Effects on Materials," and "Effects on Human Health and Visibility."

The NAPAP Final Assessment, scheduled for 1990, is currently in the planning phase. A proposed plan for the 1990 Assessment will be released for public review and comment during 1988.

RESEARCH COORDINATION

NAPAP coordination with States, regional associations, and the private sector is an ongoing process. NAPAP participated in the fifth Acid Deposition Research Coordination Workshop in October 1987, at which ten state research programs and several private research organizations were also represented. The meeting was organized by an interorganizational committee and was sponsored under a cooperative arrangement by the Electric Power Research Institute. It included reports on research activities in each of the National Program's seven research areas. Reports were delivered by NAPAP, state, and private representatives, and provided the basis for a broad, research-based dialogue. A detailed report from the meeting is available from the NAPAP Office of the Director of Research (ODR). The States have collectively appointed research representatives to facilitate coordination and communication with each Task Group. The ODR has a staff member for State liaison to further facilitate productive interactions.

Coordination is further supplemented by frequent professional contacts of ODR and Task Force staff with representatives from private sector and State research efforts. These contacts include participation in workshops and peer reviews, which provide an enhanced understanding of activities at the private, State and Federal levels and an opportunity to efficiently coordinate research activities.

The National Program has an active and ongoing tradition of research cooperation and coordination with Canada, the Federal Republic of Germany, and several other European and Scandinavian nations. Cooperative endeavors include joint projects, the sharing of data and technical information, and, in the case of the Federal Republic of Germany, the translation of important documents and technical reports. In 1987 an agreement was reached with China for the provision of atmospheric deposition data. Various research ties have also been established with Chile, Mexico, the Soviet Union, and Venezuela.

The National Program's most extensive research partnership has been with Canada. Table 1 outlines joint U.S./Canadian projects either initiated or ongoing in 1987. These projects and other research activities are reviewed each year in discussions between the NAPAP leadership and the Canadian Federal-Provincial Research and Monitoring Coordinating Committee (RMCC).

NAPAP PEER REVIEW ACTIVITIES

The quality and relevance of NAPAP research are evaluated through reviews by panels of independent experts. Peer and program reviews are conducted on a project-by-project basis as well as on a programmatic scale. These reviews help to evaluate not only the soundness of individual studies, but also the strength of the interrelationships among projects and programs. In 1987 the Terrestrial and Aquatic Effects Task Groups conducted peer reviews, and the Atmospheric Transport and Modeling Task Group held a review of its Regional Acid Deposition Model (RADM).

In all three cases the reviewers made specific recommendations for strengthening individual projects as well as groups of projects. Reports from these reviews have been widely distributed, and Task Group managers have already incorporated many of these suggestions into their research plans. Formal responses to these reviews are provided to the Interagency Science Committee.

Table 1.

U.S./CANADIAN COOPERATIVE ACID DEPOSITION RESEARCH PROJECTS

ATMOSPHERIC PROCESSES:

Activity	Purpose	Participating Organizations: U.S.	Participating Organizations: Canadian
Joint research planning and design	Eulerian acid deposition and oxidant model development	EPA, State Univ. of New York/Albany	Ontario Ministry of the Environment, Atmospheric Environment Service
Field study	Data collection for performance evaluation of acid deposition models	EPA, Electric Power Research Institute, NOAA, TVA	Ontario Ministry of the Environment, Atmospheric Environment Service
Model evaluation	Evaluation of Eulerian acidic deposition models	EPA, Electric Power Research Institute	Atmospheric Environment Service
Cooperative field study	To determine pollutant concentration, deposition and horizontal flux with distance over the North Atlantic (WATOX)	NOAA, Univ. of Va., Bermuda Biological Station	Atmospheric Environment Service
Joint research, common data base	Emissions research and national emissions inventories	EPA, DOE, NOAA	Environment Canada
Equipment transfer	Air quality/fog sampling at the Bay of Fundy and other eastern North American sites	NOAA	Environment Canada

DEPOSITION MONITORING:

Activity	Purpose	U.S.	Canadian
Network intercomparison, co-location of samplers	Intercomparison of wet deposition networks, i.e., U.S. National Trends Network (NTN) and Canadian Air and Precipitation Monitoring Network (CAPMoN)	USGS, National Atmospheric Deposition Prog., Illinois State Water Survey	Atmospheric Environment Service, Environmental Conservation Service
Laboratory intercomparison	Intercomparison of chemical methods used to analyze wet deposition samples	USGS, Illinois State Water Survey	Atmospheric Environment Service
Common data base	Unified acid deposition data base for eastern North America	EPA, USGS, National Atmospheric Deposition Prog., Pacific NW National Lab.	Ontario Ministry of the Environment, Atmospheric Environment Service
Methods research	Development of standard methods to measure dry deposition	NOAA	Atmospheric Environment Service
Network intercomparison, co-location of samplers	Intercomparison and co-location of sites for planned dry deposition monitoring networks	NOAA, EPA	Atmospheric Environment Service, Environmental Conservation Service

Table 1. (continued)

Activity	Purpose	Participating Organizations: U.S.	Participating Organizations: Canadian
DEPOSITION MONITORING:			
Monitoring	Continue to develop and expand inventory of wet deposition monitoring sites within the transboundary region	EPA, NOAA, DOE	Atmospheric Environment Service
EFFECTS ON FORESTS:			
North American Forestry Commission	Atmospheric Deposition Study Group: to coordinate research on forest effects/atmospheric deposition in the U.S., Canada and Mexico	USDA Forest Service	Forestry Service
Data exchange	Impact of acidic fog and clouds on forests	USDA Forest Service, EPA, State Univ. of New York/Albany	Forestry Service, Atmospheric Environment Service, McGill Univ.
Ongoing research	- Assess cause(s) of sugar maple decline in Northeastern U.S. and Canada	USDA Forest Service	Forestry Service
	- Assess cause(s) of decline in western conifers	USDA Forest Service	Forestry Service
AQUATIC EFFECTS:			
Monitoring program	Exchange of information on protocols and quality assurance activities	EPA	Environmental Conservation Service
Data exchange	Interpretation and evaluation of U.S. and Canadian surface water chemistry surveys	EPA	National Water Research Institute
EFFECTS ON MATERIALS:			
Co-location of samples at a variety of sites	Effects of acid deposition on the corrosion of structural materials	USGS, Bureau of Mines, EPA, Natl. Park Service	Atmospheric Environment Service
OTHER:			
Joint peer reviews	Canadian participation in Task Group research peer reviews	NAPAP Office of the Director	RMCC/LRTAP Liaison Office

In 1988, the Task Groups on Emissions and Controls, Atmospheric Chemistry, Atmospheric Modeling and Transport, Atmospheric Deposition and Air Quality Monitoring, and Effects on Materials and Cultural Resources will undergo peer/program review. Another function of peer review is to coordinate research with other organizations: recent program reviews have included presentations of Canadian research projects. In addition, this opportunity has also been provided to state and private sector research managers.

NAPAP BUDGET

For fiscal year 1987, the National Program's budget totaled $86.1 million. (See Table 2, below.) The budget was apportioned among research areas as follows: Aquatic Effects (28 percent), Terrestrial Effects (24 percent), Atmospheric Chemistry (14 percent), Atmospheric Modeling and Transport (12 percent), Atmospheric Deposition and Air Quality Monitoring (11 percent), Emissions and Controls (5 percent), Effects on Materials and Cultural Resources (4 percent), and assessment activities (2 percent). The Environmental Protection Agency provided almost 64 percent of the total, and the Departments of Agriculture, Energy, and Interior contributed 14, 10, and 6 percent, respectively. The National Oceanic and Atmospheric Administration added nearly five percent to the total, with the remaining one percent being provided by the Tennessee Valley Authority. The agency and Task Group totals and their distribution have remained relatively stable from 1986 through 1988. However, the activities of the Task Groups have changed over this period in response to progress within Task Group research programs. These research activities and their evolution are described in the seven Task Group chapters.

By working together on the Task Force, the participating agencies address national needs with an integrated and comprehensive program that avoids duplication and eliminates crucial gaps in the overall national effort.

Table 2.

NAPAP FY 1987 BUDGET

($ in thousands)

		USDA					DOI						
Task Group	EPA	CSRS	ARS	USFS	NOAA	DOE	NPS	USGS	FWS	BLM	TVA	Totals	TG. %
I	3627					750						4377	5.1%
II	4019				2806	5079					200	12104	14.1%
III	9323				940	275						10538	12.2%
IV	5420	533		146	430	806	111	1670		100	40	9256	10.7%
V	9855	2639	284	7070		40					400	20288	23.6%
VI	20012			1399		250	374	1044	947			24026	27.9%
VII	2715						825	155				3695	4.3%
Assessment						1850						1850	2.1%
		3172	284	8615			1310	2869	947	100			
Totals:	54971		12071		4176	9050		5226			640	$ 86134	100.0%

EMISSIONS AND CONTROLS

INTRODUCTION

The Emissions and Controls Task Group is responsible for providing data on past and current emissions of gases and particles that are thought to contribute to acidic deposition. The Task Group develops models which allow policy analysts to determine how policy changes affect future emissions. The Task Group also assembles data on the performance and cost of various emissions control technologies. Within the United States, sulfur dioxide (SO_2) and oxides of nitrogen (NO_x) are emitted mainly by electricity generation, transportation, and other fuel-burning processes (See Table I-1). Transportation, petroleum refining, chemical manufacturing, and paint and solvent use are the major sources of volatile organic compounds (VOC), some of which help transform sulfur dioxide and nitrogen oxides to acidic compounds in the atmosphere.

Natural sources such as soils, tidal areas, ocean waters, dust storms, and lightning discharges may also be important contributors to these emissions. Natural sources contribute significantly to total VOC emissions, and possibly to NO_x emissions. Over the eastern United States, natural source contributions to the total annual emissions of sulfur are negligible (approximately 0.1 percent). Research on natural sources is conducted by the Atmospheric Chemistry Task Group; their results will be incorporated by the Emissions and Controls Task Group into its detailed emissions data bases for use by atmospheric modelers.

1990 RESEARCH OBJECTIVES

- Assess natural and anthropogenic emissions of SO_2, NO_x, and VOC, and identify their sources:
  - Seasonally by region 1900 to 1980;
  - Monthly by state 1975 to 1989;
  - Detailed by season, typical hourly profiles for seasons, all relevant chemical species, and 20 x 20 km grid for 1980 and 1985; and
  - Natural sources by region.

TABLE I-1. Emissions by Source Category, 1980 and 1986

(10^6 metric tons/yr)

	Source	1980	1986
SO_2	Utility	15.9	14.4
	Industrial Fuel Combustion	2.4	2.2
	Comm./Institutional/Residential	0.9	0.6
	Industrial Processes	3.5	3.0
	Other	0.9	0.8
	TOTALS	23.6	21.0
NO_x	Utility	5.8	6.3
	Industrial Fuel Combustion	3.0	2.6
	Comm./Institutional/Residential	0.7	0.6
	Industrial Processes	0.7	0.6
	Transportation	7.2	7.0
	Other	2.0	2.0
	TOTALS	19.4	19.1

- Develop a comprehensive model set for predicting emissions and control costs of SO_2, NO_x, and VOC under alternative assumptions of production, technology, and economics.

- Develop detailed information on cost and performance of existing and potential control technologies and energy technologies.

RESEARCH ACTIVITIES IN 1987

Emission Trends/Detailed 1985 Emission Inventory

The historic (1900 to 1980) NO_x/SO_x emissions data base was expanded to include VOC emissions. Efforts were also undertaken to reconcile this data base with other NO_x/SO_x estimates, such as those of the National Academy of Sciences.

The current emissions trends data base (i.e., the Monthly/State NO_x/SO_x 1975 - Present Data Base) was updated to include 1985 and 1986 emissions and also to add VOC emissions. Initial estimates of uncertainties for the SO_2 and NO_x data bases were developed from statistical analyses. The methodologies used in these uncertainty analyses will also be available for use in estimating the uncertainties in the other emissions data bases.

For the 1985 detailed emission inventory, extensive checking of state inputs continued and further attempts to resolve problems were undertaken. All of the point source data were then returned to the states for review and comment. The area source emission component of the inventory was developed and sent out for state review. Work also began to incorporate emissions data from Canada.

Integrated Emissions Projection Model Set

The entire integrated Advanced Utility Simulation Model (AUSM) was completed, tested, and evaluated. Its forecasts were compared with those from the proprietary Coal and Electric Utilities Model maintained by ICF, Inc., using a similar set of parameters. The emissions forecasts from each model were almost identical. Completion of this publicly available information which has produced forecasts equivalent to a highly developed mature model is a major accomplishment of this program.

The Industrial Combustion Emissions (ICE) Model underwent additional testing, and final modifications to the decision logic for the fuel choice module were documented. The Process Model Projection Technique (PROMPT) and Volatile Organic Compound Model were documented and underwent further sensitivity testing along with the Transportation and Commercial/Residential Models (TEEMS and CRESS). Contractors were selected for detailed external reviews of TEEMS, CRESS, the Energy/Economic Driver, and the Integrated Emissions Projection Model. Work began on assembling the Integrated Emissions Model Set.

Control Technology Studies

Detailed site and performance information were obtained for 12 coal-fired power plants in Ohio, Kentucky, Tennessee, and Alabama. A draft report on the feasibility and cost of existing and emerging SO_2 and NO_x control technologies at these sites was completed and issued for peer review. In addition, several reports on evaluation of emission control technologies, including "generic" cost estimates, were completed and issued for peer review.

FUTURE RESEARCH ACTIVITY

Emission Trends

No further work is planned on the Historic NO_x/SO_x/VOC 1900-1980 Emissions Data Base. Final reports will be published in early 1988. Annual updates of the Monthly/State 1975 - Present NO_x/SO_x/VOC data base will continue to the end of the program with a major reconciliation with the NAPAP 1985 Detailed Emission Inventory planned for 1989.

1985 Detailed Emissions Inventory

Work on the development of this highly detailed emissions data base will continue. The focus of the effort in 1988 will be to allocate the major point source and county-area source emissions estimates of criteria pollutants which have been assembled into the detailed and speciated data needed as input to the NAPAP atmospheric transformation and transport models such as the Regional Acid Deposition Model (RADM). These models require hourly input estimates of some 20 to 25 chemical species into approximately 20 x 20 km grids for all of the United States and parts of Canada.

Estimates of uncertainties will accompany final derived quantities. This detailed inventory will provide a basis for assessment and regulatory analysis, basic input to the application of RADM and other atmospheric models, evaluation tests of the RADM, and calibration of the monthly/state operational trend inventory. The inventory will be completed in FY 89.

Integrated Emissions Projection Model Set

Development of the Integrated Model Set will be complete in 1988. It will be peer reviewed, modified, tested, and fully documented during FY 88 and FY 89 and applied in FY 90 to examine relevant emission control scenarios.

Control Technology Studies

Efforts in 1989 will be directed toward (1) completing the site-specific SO_2 and NO_x performance/cost retrofit studies of the top 200 coal-fired utility plants (in terms of SO_2 emissions) in the eastern United States and (2) updating the status of current and advanced technologies for control of SO_2, NO_x, and VOC emissions. In the update, estimates of capital and annual costs, percent emissions reduction, and cost per unit of pollutant removed will be reported for both new sources and retrofit situations. Technology developments in the United States and abroad will be included in the update.

1987 RESEARCH HIGHLIGHTS

- The completion of the historical inventory for SO_2, NO_x, and VOC (1900 to 1980) provides state/annual data for use in the final NAPAP Assessment.

- The methodology used in developing the Monthly/State NO_x/SO_x/VOC 1975 - Present current emissions trends was modified so that initial estimates of the previous year's emissions could be made in the first quarter of each year. Using these methodology improvements, estimates were made in 1987 for emissions in both 1985 and 1986.

- The completion of the point source review for the 1985 detailed emission inventory has provided a much improved data set over the 1980 data base. Users of the 1985 data base for the RADM atmospheric transport/transformation model will have greater confidence in the emission estimates as a result of the error-checking procedures employed in reviewing the data base.

- The completion and sensitivity testing of all of the models of the Integrated Emissions Model Set provided valuable information on how the individual models perform and how they are likely to fit together when assembled into the fully integrated system. Third-party review will provide further improvements in the individual models.

- Development of the Advanced Utility Simulation Model (AUSM) was completed. Control technology costs for sorbent injection (LIMB) were modified and incorporated into the pollution control model within AUSM. Calculation of dividends and the cost of capital in the financial calculation model within AUSM were altered, and changes to reflect current tax laws incorporated. The Unit Inventory data base within AUSM for the year 1980 was updated, and will be upgraded to 1985 data in 1988. Extensive comparisons were made with ICF's Coal Electricity Use Model (CEUM) for a common set of economic forecast input assumptions. Outputs from each model were similar. The second phase of scientific peer review of the national-level version of AUSM was also conducted by the Energy Laboratory of the Massachusetts Institute of Technology.

- The completion of 12 detailed site studies at coal-fired power plants has reduced the uncertainty regarding retrofit difficulty factors and costs for a wide variety of site conditions and boiler types. These data have been used to develop simplified procedures for evaluating the top 200 coal-fired power plants (in terms of SO_2 emissions) in the eastern United States. These procedures will provide the basis for a more accurate estimate of control technology retrofit difficulty and costs for major utility sources of SO_2 and NO_x.

MAJOR DELIVERABLES

Deliverable	Date	Significance
Detailed Emissions Inventory:		Provides emissions of approximately 26 species to 20-km grid and hourly profiles as input to the atmospheric transport/transformation model (RADM). Also provides source identification for policy analysis.
- Data bases and codes	1989	
- Summary reports	1989	
- Uncertainty reports	1989	
- Annual update: utility file	1989, 90	
Current Emission Trends:		Provides state/monthly data on NO_x, SO_2, and VOC emissions for use with current deposition/concentration data in developing and analyzing source/receptor hypotheses.
- 1975 to Present		
- Yearly Update		
- Flash report	1989, 90	
- Final report	1989, 90	
Integrated Emissions Projection Model Set:		Provides a tool for estimating state-level emissions of NO_x, SO_2, and VOC to the year 2030 as a function of socioeconomic input, and computes the cost of generic control scenarios.
- Data bases and codes	1989	
- Validation report	1990	
- User's manual	1990	
Generic SO_2/NO_x/VOC Control Technology and Costs:		Provides information on the cost, applicability, and control level of new and emerging SO_2, NO_x, and VOC control technologies for use in assessment.
- Final report	1990	
Site-Specific SO_2 and NO_x Control Costs:		Provides site-specific information on retrofit difficulty and costs for existing and emerging SO_2 and NO_x control technologies applied to the 200 top emitting coal-fired plants in the eastern United States.
- Final report	1989	
Eastern Low-Sulfur Coal Reserve Study:		Provides more accurate and updated information on bed depths, thicknessess, sulfur content and applicable mining methods for low-sulfur coal reserves for use in the Advanced Utility Simulation Model (AUSM) to determine availability and mining cost for these reserves.
- Final report	1990	
Comparison of Technology Costs:		Provides a comparison of relative costs of competing coal-fired electric generating alternatives (conventional, new units, life extension, repowering and reboilering).
- Final report	1989	

ATMOSPHERIC CHEMISTRY

INTRODUCTION

The Atmospheric Chemistry Task Group is concerned with factors that modify the chemical composition of the air during transport from points of emission to the vicinity of receptors. These factors include: (a) surface emissions due to natural sources, (b) chemical reactions that occur in clear air, (c) reactions and scavenging associated with clouds, (d) interception of cloud liquid water by forests (especially at mountain tops), and (e) dry deposition. Subtask groups are organized accordingly. In each case, the focus is on the identification of critical processes, and their characterization both for inclusion in transport models and for improving estimates of deposition from limited field data.

1990 RESEARCH OBJECTIVES

- To quantify the natural background flux of sulfur, nitrogen oxides (NO_x), ammonia (NH_3), volatile organic compounds (VOC), and alkaline dust into air and provide the basis for estimating background precipitation chemistry and air quality on a regional basis by season.

- To produce an adequate understanding of chemical processes in clear air that produce acids and related oxidants from the precursors sulfur dioxide (SO_2), NO_x, and VOC.

- To produce an adequate understanding of chemical reactions in the aqueous phase of clouds, fog, and precipitation, the scavenging of acidic species in vapor or particulates from the atmosphere, and transport processes associated with clouds and storms.

- To establish the concentration of acidic species and oxidants in mountain clouds and ground-level fogs.

- To develop quantitative measurement systems for dry deposition, and use these to determine and formulate the processes causing dry deposition. To develop routine monitoring systems based on the results of these studies, and to deploy these systems in trial monitoring programs.

The five distinct sub-task groups share the common objective to develop and test formulations, descriptions, and computer modules that can be linked effectively into acid deposition models being developed by the Atmospheric Transport and Modeling Task Group or which will serve to improve the estimates of deposition and exposure regimes required by Terrestrial, Aquatic, and Materials Effects Task Groups.

RESEARCH ACTIVITIES IN 1987

Natural Emissions

Natural (mostly biogenic) and anthropogenic emissions of nitrogen and sulfur compounds combine in the atmosphere with volatile organic compounds, leading to the deposition of acid nitrate, acid sulfate, and oxidants. To account for the role of the natural sources, studies of related natural emissions were made to: 1) develop techniques to measure the critical chemical species at low concentration levels, 2) use these techniques to measure atmospheric concentrations at remote sites to establish upper limits on the natural background, 3) critically assess the techniques used to determine fluxes from soils, 4) use these techniques to measure emissions from soils at representative sites in North America, and 5) provide a national inventory of related emissions from natural sources.

During 1987, studies conducted in Pennsylvania and Colorado used enclosure and gradient techniques to quantify NO_x emission rates from the soil. Emission rates were formulated in terms of atmospheric and soil properties. These relationships are the basis for re-assessing the relative importance of natural NO_x emissions in the eastern United States. Similar field studies were carried out for the emissions of natural VOC.

Clear Air Chemistry

The program on clear air chemistry is designed to provide laboratory data and field measurements needed to assess the rates and mechanisms of acid formation in clear air. Recent work has focused on two issues: (1) the production of SO_2 from reduced sulfur compounds derived from natural sources and (2) the credibility of field measurements of atmospheric trace gases. The first issue is related to the need to specify the baseline atmospheric concentrations of sulfur compounds due to natural emissions. The second concern remains a central issue. Measurements of nitrogen oxides are of special interest, both because NO_x is important in producing acid deposition and a variety of oxidants, and because it is difficult to measure.

Aqueous Chemistry and Scavenging

During June 1987, a major field study of precipitation scavenging was conducted. This study, "PRECP V", the fifth in the sequence of PRocessing of Emissions by Clouds and Precipitation (PRECP) field experiments, was held in the vicinity of Columbus, Ohio, and addressed two primary objectives: (1) the role of the vertical distribution of hydrogen peroxide on in-storm SO_2 oxidation and scavenging, and (2) the quantification of cloud venting, whereby pollutants in the cloud are emitted into the free troposphere aloft. The first issue is of considerable importance because hydrogen peroxide (H_2O_2) is now known to be the dominant oxidizing agent by which SO_2 is converted to sulfuric acid (H_2SO_4) in many clouds, but the distribution of H_2O_2 is not yet known. The second focus, cloud venting, is of special interest in the context of long-range transport. Once pollutants are transported into the upper atmosphere, they can be carried for long distances before being scavenged or deposited. Extensive laboratory investigations have continued in parallel with the field experiments. Recent laboratory work has concentrated on the measurement and formulation of the uptake of pollutant molecules by water droplets.

Mountain Cloud and Fog Chemistry

This program is designed to assess the exposure of high-elevation forests in eastern North America to clouds and associated atmospheric chemicals. In 1987, three intensive measurement periods were conducted during the forest growing season (April to October). Parameters measured included cloud frequency, chemical concentrations in cloud liquid water, wet and dry deposition, ozone, sulfur dioxide, hydrogen peroxide and a range of supporting meteorological properties.

Dry Deposition Processes

Operations at the three Core Research Establishment (CORE) sites and at the array of simpler satellite locations have continued. Specific attention has been directed to the relative importance of wet and dry deposition, primarily for sulfur and nitrogen species, and to particle deposition. Improvement of measurement techniques continued to be a major activity. In conjunction with the Terrestrial Effects Task Group, experiments have been conducted at Walker Branch Watershed, Tennessee, with S-35 (which has an 87-day half-life) to quantify the contribution of foliar leaching and dry deposition to sulfate ($SO_4^=$) in net throughfall. The measurement of deposition by throughfall has been extended to an array of measurement sites in the southern Appalachian Mountains. The high-elevation site is in a spruce forest at an elevation of 1800 m in the Great Smoky Mountains National Park, and the low-elevation site is in a pine forest at 300 m in the foothills of the Cumberland Plateau, 90 km to the northwest. These sites are part of a larger-scale study of deposition effects on forest nutrient cycling.

FUTURE RESEARCH ACTIVITY

Natural Emissions

Additional measurements will be made to evaluate NO_x emission estimates in representative biomes in the United States. If the representativeness of these estimates can be established, then the underlying algorithm will be used as the basis for a spatially and seasonally resolved national inventory of natural NO_x emissions.

In order to understand regional oxidant formation and acid transformation processes, the relative contributions of natural and anthropogenic sources to regional levels of VOC will be investigated further, along with the reactivities of individual compounds or classes. This will involve studies of natural VOC emissions from various biomes, along with measurements of the ambient concentrations of natural and man-made VOC in the nonurban atmosphere.

Clear Air Chemistry

Future work will concentrate on two areas of research: (1) the removal of gaseous species on surfaces and (2) the further investigation of oxidation mechanisms for atmospheric anthropogenic sulfur and nitrogen compounds. Further comparisons are planned to evaluate NO_2 measuring techniques. The measurement of VOC will also be investigated. Current comparisons show that, while present techniques are probably satisfactory for simple hydrocarbons, more sophisticated methods are required to measure heavier hydrocarbons and partially oxygenated VOC.

Aqueous Chemistry and Scavenging

The sequence of field studies will continue to focus on the mechanisms by which convective clouds scavenge, transform and deposit airborne pollutants, with increasing emphasis on cloud venting (by which pollutants are relocated to greater altitudes). The sixth field study of the PRECP series will be an investigation of convective storms and their scavenging characteristics, to be conducted in central Illinois in the spring of 1988. The study is designed to identify and quantify dynamic, microphysical, and chemical processes that influence precipitation scavenging in convective clouds, and to evaluate convective components of scavenging modules for regional models. The field experiments of the PRECP series will culminate in a sequence of tests of the wet deposition predictions of the Regional Acid Deposition Model for frontal storm situations, tentatively proposed for Pennsylvania and southern Ontario.

Mountain Cloud and Fog Chemistry

Emphasis will continue to focus on assessing the above-cloudbase environment to which forest ecosystems are exposed. Research will concentrate on three main subjects: 1) the exposure of high-altitude forests to gaseous air pollutants and clouds, 2) the relative significance of deposition mechanisms in mountainous forests, and 3) the variation in deposition of air pollutants as a function of elevation. Individual sites of the Mountain Cloud and Fog Chemistry network will continue to investigate aspects of exchange mechanisms that are amenable to study. Several sites will employ throughfall methods to quantify deposition, whereas others will use micrometeorological techniques.

Dry Deposition Processes

Activities at the various dry deposition research sites will focus increasingly on the parameterization of processes controlling uptake at surfaces, especially stomatal resistance. NO_x exchange and particle deposition remain especially uncertain. Studies will be conducted to test existing deposition velocity formulations in demanding circumstances. Comparisons will be made between the predictions of deposition velocity formulations developed for regional-scale models (e.g., RADM) and the routines developed for interpreting data obtained in measurement networks (the NOAA "Big Leaf" model). Exploratory studies will be conducted of the representativeness of localized dry deposition measurements. The question that arises is how to extrapolate from site-specific information to model grid cells, and how to make use of model outputs when the target area of interest is far smaller than a model grid cell.

1987 RESEARCH HIGHLIGHTS

- The importance of natural sulfur emissions relative to anthropogenic sources in the eastern United States has been re-examined. Emissions of sulfur gases from natural land surfaces have been estimated. For the eastern U. S., the result (about 13,000 metric tons of sulfur per year) is only about 0.1 percent of the reported annual emission of anthropogenic sulfur. This is more than twenty times less than an earlier upper-bound estimate based on preliminary data.

- Results of studies in Pennsylvania emphasized the influence of natural emissions, primarily of VOC, on the production of ozone at a rural site in the East. Most ozone in the lower atmosphere is produced through photochemical reactions that involve simultaneously both NO_x and VOC as the major precursors. Previous studies have established that anthropogenic emissions are the major source of NO_x over rural areas in the eastern United States. However, the importance of natural VOC relative to anthropogenic VOC has remained a subject of debate.

- A theoretical photochemical model was used to analyze measurements of ozone, NO_x, and natural and anthropogenic VOC obtained in the 1986 field study in Pennsylvania. The results are illustrated in Figure II-1. The diagram shows the model-calculated ozone buildup over a 4-day period (Figure II-1a) when concentrations of observed isoprene (a natural VOC) are added (curve 2) to a background environment (curve 1) that contains only observed levels of methane, carbon monoxide and NO_x. The calculated increase in ozone production is substantial. When observed anthropogenic VOC are added to the natural VOC (curve 3), the calculated additional ozone buildup is much smaller. The resulting calculated total ozone levels match the values observed (Figure II-1b), during much of this modeled episode, indicating that the model is adequately representing the chemistry at this time and place. The key feature is the relatively small calculated ozone increase due to addition of the anthropogenic VOC; this effect occurs primarily because the ozone production rate becomes nonlinear at the higher total VOC levels for a given NO_x concentration (i.e., there is less ozone production for the same VOC increase). Similar results would be obtained if anthropogenic VOC were first reacted with the NO_x and isoprene added later, such as in urban and suburban areas. However, it must be stressed that these results were obtained during one summer at a single location. Experiments aimed at testing these preliminary conclusions are planned for 1988.

- The oxidation mechanisms for several reduced sulfur compounds derived from natural sources have been investigated. These processes result in the production of SO_2 throughout the atmosphere. Some sulfur species react with hydroxyl radicals in the lower atmosphere; others are relatively inert in the lower atmosphere and are transported into the stratosphere before oxidation takes place. Studies of the gas-phase oxidation of SO_2 to H_2SO_4 indicated that sulfur trioxide is formed as an intermediate in the oxidation.

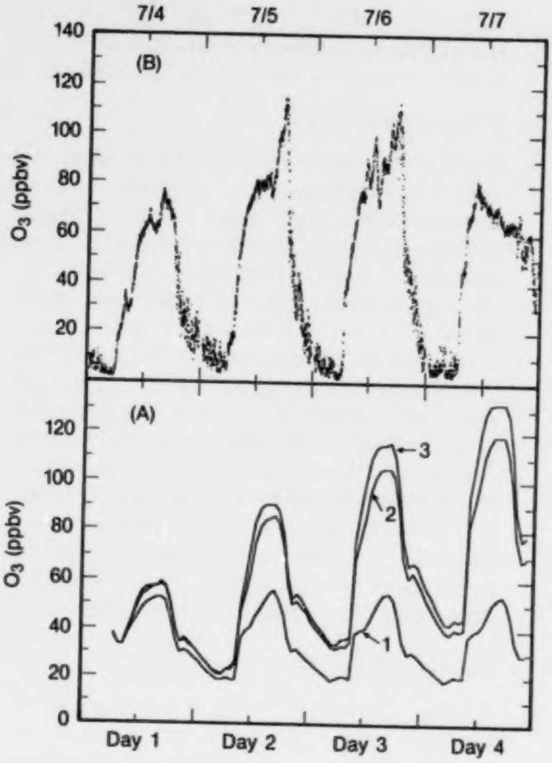

Figure II-1. (A) The model-calculated variation of ozone concentration during a four-day period assuming (1) CO and CH_4 mechanisms only, (2) an additional production of ozone due to isoprene, and (3) a further additional role for anthropogenic volatile organic compounds. In each case, the measured NO_x concentrations were used in the computation. (B) The corresponding variation of ozone concentrations observed during the Pennsylvania study.

- Systematic errors were detected in the measurement of nitrogen oxides by some techniques in common use. Surface-conversion methods (e.g., ferrous sulfate and molybdenum) systematically overestimate NO_2 concentrations.

- One of the principal issues at the outset of PRECP was the possible non-linear relationship between emissions and deposition in eastern North America. Important sources of this non linearity include the limited availability of hydrogen peroxide as an oxidant of sulfur dioxide and inhibition of ozone-induced oxidation as a consequence of the acidity produced. PRECP field projects have provided evidence for the occurrence of both of these mechanisms. The extent of in-cloud oxidation of sulfur dioxide by hydrogen peroxide depends on the amount of hydrogen peroxide available in-cloud relative to sulfur dioxide. Hydrogen peroxide concentrations vary with season because the dominant source of hydrogen peroxide is photochemical reactions in the atmosphere. Hydrogen peroxide concentrations can be relatively high in summer (up to several ppb), sometimes greater, sometimes less than sulfur dioxide concentrations. In winter, hydrogen peroxide concentrations are typically quite low; they have seldom been found to be greater than 0.1 ppb, and are typically much less (<0.01 to 0.05 ppb gas-phase equivalent inferred from cloud water concentrations). With respect to oxidation by ozone, although ozone concentrations are typically one to two orders of magnitude greater than sulfur dioxide concentrations, oxidation of SO_2 by O_3 occurs only to a limited extent, and the fraction of SO_2 oxidized by O_3 decreases with increasing SO_2 concentration. Consequently, so long as in-cloud oxidation of sulfur dioxide dominates over clear-air oxidation (which is frequently the case), then whenever the hydrogen peroxide concentration is less than that of sulfur dioxide (such as in winter), little of this "excess" sulfur dioxide will be oxidized within clouds and little of this sulfur dioxide will be removed from the cloud in precipitation. That is, cloud and storm systems are frequently "chemically saturated" in terms of the amount of sulfur dioxide that can be scavenged by precipitation. To the extent that decreases in sulfur dioxide emissions remove only some of this "excess" sulfur, there will then be less than proportionate change in the wet deposition of sulfate, and the source-receptor relation will display nonlinearities.

- Significant annual and latitudinal variations have been found in hydrogen peroxide concentrations in air and in cloud liquid water measured at mountaintop sites. Hydrogen peroxide (H_2O_2) is a compound of considerable interest because it may affect tree growth and because of its role in the oxidation of SO_2 in cloud water. A comparison of gas phase H_20_2 levels at Whitetop and Whiteface for the summer of 1986 is shown in Figure II-2. The diagram provides preliminary evidence that the entire probability distribution is shifted towards lower concentration values at Whiteface than at Whitetop; average levels were lower at Whiteface than at Whitetop (0.6 versus 0.8 ppb). This difference is consistent with current understanding of gas-phase H_20_2 formation which predicts higher formation rates for the higher light intensity and water vapor levels typically found in the South.

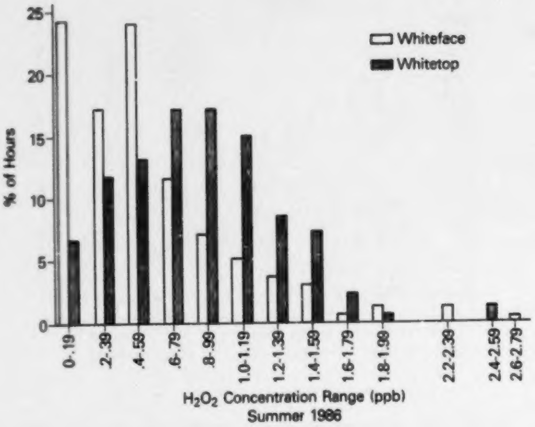

Figure II-2. A preliminary comparison of the frequency distributions of the range of hourly average gas-phase H_2O_2 concentrations, as measured during summer, 1986, at mountaintops in northern New York (Whiteface) and southern Virginia (Whitetop).

- First estimates of wet/dry deposition ratios have been produced on a network basis, as a product of the research "CORE" network and its array of simpler satellite sites. The relative importance of wet and dry deposition at the Argonne, Illinois, CORE site is illustrated for sulfur and nitrogen species in Figures II-3 and II-4. The results are based on weekly NADP/NTN collections of precipitation and the NOAA weekly filterpack measurements of concentrations in air. For total sulfur (expressed as sulfate), wet deposition is seen to exceed dry deposition in the autumn of both years, whereas the two contributions were about equal in spring and summer. For winter, the two years differed greatly; this appears to have been due primarily to the variation of wet deposition, which provided 7 mg/m^2 of total sulfur during the first winter and nearly 50 mg/m^2 during the second. This variability in wintertime deposition is also evident in the relative nitrate deposition. At other times of the year, nitrate deposition tended to be equally divided between wet and dry components. For the entire study period, 43 percent of the total sulfur deposition and 51 percent of the total nitrate deposition were by dry processes. Throughfall measurements obtained so far at an array of sites in the southern Appalachian Mountains indicate that deposition increases with elevation, and that dry and cloudwater deposition, and higher precipitation, are primarily responsible for the deposition enhancement in the mountains.

- Exploratory isotope studies of the leaching of sulfur from the interior tissue of leaves by the

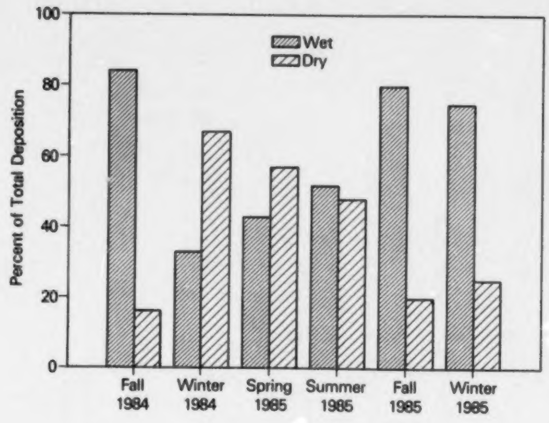

Figure II-3. A comparison of seasonal average deposition amounts of sulfate, for Argonne, Illinois, deposited as $SO_4^=$ in wet deposition and as the total of particulate sulfate and sulfur dioxide (SO_2) for the dry deposition component.

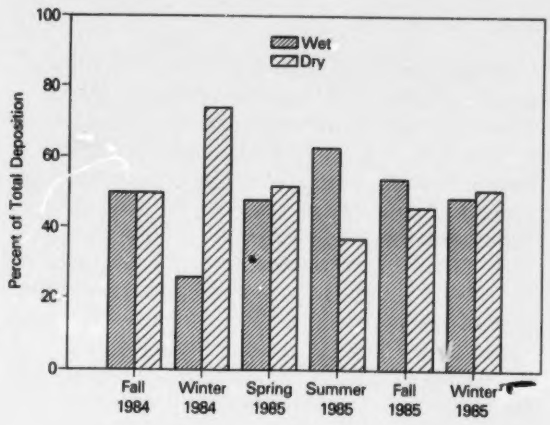

Figure II-4. A comparison of seasonal average deposition amounts of nitrate, for Argonne, Illinois, deposited as NO_3^- in wet deposition and as the total of particulate nitrate and nitric acid vapor (HNO_3) for the dry deposition component.

washing of the leaf surface have been conducted at Walker Branch Watershed, Tennessee, using S-35 (87-day half-life) to quantify the contribution of foliar leaching and dry deposition to sulfate ($SO_4^=$) in net throughfall (i.e., excluding the component directly associated with precipitation). Two red maple and two yellow poplar trees (12 to 15 m tall) were studied. It was found that foliar leaching accounted for less than 20 percent of the sulfate measured in net throughfall, (during the growing season) and dry deposition accounted for more than 80 percent. Hence, dry deposition of particles to these tree species can be reasonably approximated during summer from the measurement of $SO_4^=$ flux in net throughfall.

MAJOR DELIVERABLES

Deliverable	Date	Significance
Provision of data for determining background in North America.	1988	Background pH levels must be known, and will be derived using models precipitation pH driven by emission data.
Improved estimates of the inventory of natural hydrocarbon emissions.	1988	Natural sources are suspected to be important in the production of hydrogen peroxide.
Improved estimates of alkaline aerosol fluxes.	1988	Alkaline aerosols reduce the acidity of rain.
Assessment of the role of oceanic sulfur as a source of precipitation acidity in the Pacific Northwest.	1988	In the Northwest, sulfur in rain is derived mainly from oceanic sources.
Improvements in measurement of reactive nitrogen oxide species and ammonia for the eastern United States.	1988	The distribution of nitrogen among many different species remains difficult to measure. The speciation must be known before reactions can be predicted.

Deliverable	Date	Significance
Measurements of reactions and solubilities of ozone and other oxidants at ice surfaces.	1988	These reactions contribute substantially to the oxidation of sulfur dioxide.
Improved models of hydrogen peroxide production.	1988	Hydrogen peroxide is the key species for oxidizing sulfur dioxide to sulfate.
Improved specification of aqueous pathways for hydrogen peroxide formation.	1988	Hydrogen peroxide is the key species for oxidizing sulfur dioxide to sulfate in droplets as well as in air.
Report on the role of organics in aqueous-phase production of acids.	1988	Organic are known to be involved in the production of acids, but their relative importance remains unknown.
Complete diagnostic analysis of PRECP I-V field studies.	1988	These field data are the basis for characterizing scavenging processes.
Conduct the PRECP VI field experiment.	1988	This is a key study of scavenging and venting by convective storms.
Determination of ice-phase solubilities and reaction rates of hydrogen peroxide, nitric acid, and sulfur dioxide.	1988	In-cloud reactions and scavenging involving the ice phase must be better known to evaluate errors in RADM scavenging formulations.
Preliminary report on elevational gradients at Mountain Cloud Chemistry sites.	1988	This information is needed to assess effects under the Terrestrial Effects Task Group.
Initial assessment of the relative importance of wet, dry, and occult deposition in mountain environments.	1988	Unique conditions occur at high elevations. These data are provided for the Terrestrial Effects Task Group.
Documentation of frequency distribution describing atmospheric exposure regimes in mountain environments.	1988	This information is needed to assess effects under the Terrestrial Effects Task Group.
Report on improved methods for dry deposition measurement.	1988	Errors in dry deposition estimates due to imperfect measurement techniques must be quantified.
Assess the watershed mass balance approach to derive dry deposition rates.	1988	Methods are required to quantify dry deposition to watersheds. Information is needed for the Aquatic Effects Task Group.
Conduct an interim comparison of methods used at various research sites.	1988	Errors in dry deposition estimates due to imperfect measurement techniques must be quantified.

Deliverable	Date	Significance
Benchmark dry deposition at CORE sites to reference monitoring networks for 1987.	1988	Methods for deriving dry deposition estimates from routine observations are updated annually.
Comparison of simulated and measured cloud water composition and deposition at Whiteface Mtn., NY, and Shenandoah National Park, VA.	1989	Such tests are required to test cloud chemistry and scavenging modules, as in the RADM.
Complete annual summary of measured chemical exposure and deposition.	1989	Thorough documentation of exposure environments is required to complete the assessment of effects on high-elevation forests.
Improved inventory of nitrogen emissions from soils.	1989	Natural sources of nitrogen oxides are known to be significant. They must be quantified to specify background conditions.
Initial assessment of uncertainties associated with aerial extrapolation.	1989	Relates results from measurements at specific locations to area averages as produced by RADM.

ATMOSPHERIC MODELING AND TRANSPORT

INTRODUCTION

The primary goal of the National Program's Atmospheric Modeling and Transport Task Group is to model the contribution of emissions from sources in one region to deposition at receptors in another (source attribution), and to model the change in deposition at those receptors accompanying changes in emissions. Such efforts entail the credible and efficient quantification of the complex dynamics associated with atmospheric transport, transformation, and deposition.

Model development must be followed by field tests of the models (model evaluation) to assess their performance by comparing predictions to observations. The application of the new models to answer scientific questions and provide deposition data for effects research is the primary means for the National Program to reduce uncertainties in source attribution and receptor response. Furthermore, special studies are required to obtain empirical data to further our understanding of the complexities of source attribution. The Task Group research objectives evolved from this program perspective.

1990 RESEARCH OBJECTIVES

- Provide scientifically valid and efficient source/receptor models on both the regional scale and the mesoscale that are fully documented.
- Complete evaluation of these models to determine their level of accuracy and reliability.
- Demonstrate the application of these models to:
  - Predict changes in deposition and air quality at sensitive areas from given changes in emissions from specified sources,
  - Establish relative importance of local vs. distant sources, and
  - Examine non-linearities in source/receptor relationships.
- Investigate characteristics of regional transport with respect to (a) meteorological and chemical influences on deposition, (b) local influences of major emission sources, and (c) transport of species across political boundaries.

RESEARCH ACTIVITIES IN 1987

Model Development

The developmental phase for the Regional Acid Deposition Model (RADM) was completed in 1987. As the centerpiece of this effort, a major peer review of the RADM program was held in May 1987 and recommendations were used to improve the National Program's atmospheric modeling program. The development of Meso-STEM, an Eulerian acid deposition research model for the urban scale, was completed.

Model Evaluation

Model evaluation activities were centered on preparations for the upcoming field study to collect data for use in the evaluation of the RADM model. Field study preparations included design studies which used data and outputs from the RADM and the Engineering Model.* Design workshops were held to further define the use of aircraft in the field study. The preliminary evaluation of RADM against available data bases continued, with an examination of the effect of reducing errors in input meteorology on evaluation results.

Model Application

An application procedure, termed aggregation, which will enable annual average deposition to be estimated from a subset of episodes, was designed for use with the RADM model. Preliminary work began to provide model-based dry deposition estimates for use by the Aquatics Effects Task Group. Aspects of nonlinearity were explored using the RADM model and the Engineering Model. An emissions sensitivity study was developed to aid in the identification of crucial data to be collected during the field study to reduce SO_2 emissions uncertainty.

Special Studies:
Characteristics of Regional Transport

Several special studies research projects begun in 1985 and 1986 have been completed. The Washington, DC urban deposition measurement study was conducted as a follow-on of a Philadelphia study designed to assess the local contribution of urban areas to wet deposition. The ASARCO Study (the ASARCO Smelter, Tacoma, Washington), which looked for and detected the local influence of a major point source of SO_2 emissions, was completed. The Western Atlantic Ocean Experiment (WATOX) involved the measurement of the flux of a variety of gases and aerosols off the east coast of North America from Florida to Newfoundland. Data collection for the Across North American Tracer Experiment (ANATEX), a follow-on to the Cross-Appalachian Tracer Experiment (CAPTEX), including more complex meteorology, was completed and data analysis is being undertaken.

FUTURE RESEARCH ACTIVITY

There will be a major shift in this Task Group's emphasis in 1988. Work and resources will shift from model development to model evaluation and application. While development of the MesoSTEM model was completed in 1987, the completion and documentation of the NAPAP applications version of RADM** will occur in 1988. A high priority will be given to carrying out the new field study to support model evaluation needs. Priority will also be given to completing modeling tools and methods to support NAPAP applications work. Also, preliminary analyses will begin which concern questions associated with linking emissions from one region to deposition in another.

1987 RESEARCH HIGHLIGHTS

- The Pacific Northwest Laboratory (PNL) and the University of Washington studied the effect on precipitation acidity due to the shutdown of the ASARCO copper smelter in March 1985. The study was conducted by collecting rain samples in the Puget Sound area both before and after the smelter was shut down.

  Researchers found that the smelter was responsible for approximately half of the sulfate deposition in the downwind rain. The area of significant effect occurred over 600 km^2 and extended 25 km downwind of the source. Further downwind, over Seattle, an effect was observed, but it was not statistically significant. For the seven storms sampled when the smelter was operating, the wet deposition of sulfate averaged 0.022 kg/ha/event, representing approximately 1.7 percent of the smelter emissions. Thus, even though only a small fraction of the sulfur emissions are transformed into sulfate and deposited in rain over the 600-km^2 downwind area, this accounts for about half of the sulfate deposited in the rain in this area. A further fraction of the sulfur emissions will be dry deposited within the 600-km^2 downwind area, and the remaining amount will be diffused through the atmosphere and deposited in both dry and wet forms over a much wider area.

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* The Engineering Model is a simplified version of the Regional Acid Deposition Model (RADM), designed to run on a VAX computer. This model is designed to permit rapid application tests without sacrificing the fundamental description of the chemical and physical processes.

** RADM II, the second-generation Eulerian acid deposition model under development which will incorporate advanced scavenging and dry deposition modules. The RADM II will also feature a simplified code which will increase its speed by a factor of two over RADM I.

- During January 1986, a series of research flights was conducted over the Western Atlantic Ocean, 200-300 km off the coast of North America at altitudes up to 5.5 km. Flights were made parallel to the coast during periods of northwesterly flow to establish the continent-to-ocean flux of gases and aerosols in the marine boundary layer as well as in the free troposphere. Measurements were made of a variety of gases including sulfur dioxide (SO_2), nitrogen oxides (NO_x), and ozone (O_3). In addition, aerosols were collected and analyzed.

Analysis of these gas and aerosol measurements has yielded several important conclusions. Specifically, ozone profiles in the free troposphere typically indicated concentrations in the 30 to 40 ppb range, except at the base of clouds which are associated with a strong front where concentrations are as high as 70 ppb; concentrations in the marine boundary layer tended to be lower than in the free troposphere. These values are important because they confirm elevated ozone levels far (over 200 km) from major anthropogenic sources. Throughout the measurement period, many of the gas and aerosol species were highly correlated, indicating that these materials were transported as parcels. Perhaps most importantly, this research program allowed the sulfur and nitrogen budgets for the Western North Atlantic to be refined. Past and current data indicate that, on an annual basis, approximately 25 to 35 percent of the sulfur oxides and 15 to 25 percent of the nitrogen oxides emitted to the eastern North American atmosphere are transported out from the continent over the Atlantic Ocean.

- The Central U.S. RADM Test and Assessment Intensives (CURTAIN) is a series of atmospheric sampling flights providing observations for the determination of western boundary conditions for the Regional Acid Deposition Model (RADM). The experiment used identical flight plans along the Mississippi River between Iowa and the Gulf of Mexico during one month in each of the four seasons in 1987. The data set included continuous measures of trace gases (O_3, SO_2, hydrogen peroxide [H_2O_2], and NO_x), meteorological variables, position, and aerosol numbers and sizes. Summary data for hydrogen peroxide (H_2O_2) and ozone (O_3) are shown in Figure III-1. The seasonal variation of both hydrogen peroxide and ozone agree with those found elsewhere. In addition, the concentration of hydrogen peroxide was found to be higher over southerly latitudes than at the more northerly latitudes.

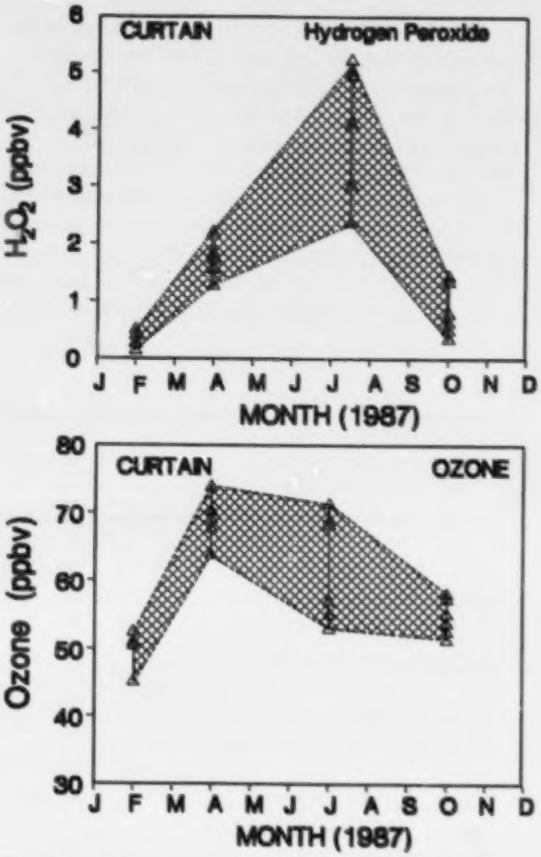

Figure III-1. Mean concentration of gas phase hydrogen peroxide (H_2O_2) and ozone (O_3) observed during each of the CURTAIN flights. Hydrogen peroxide exhibited the strongest seasonal trend, increasing by a factor of 10 or more from winter to summer. Ozone also had a distinct seasonal trend, peaking in spring and summer. CURTAIN observations comprise an accurate and complete data set, documenting the chemical state of the atmosphere above the Mississippi River valley during the four seasons of 1987. The symbol ▲ indicates mean concentrations during constant altitude (1500 or 2500 meters) portions of each flight. (Boatman et al.)

- As a first step in the evaluation of the ability of RADM to simulate measurements of deposition, RADM predictions were compared to observations made in three episodes of the Oxidation and Scavenging Characteristics of April Rains (OSCAR) program (OSCAR-I, -II, and -IV). The chemical concentrations in rainfall and deposition of sulfate and nitrate, as well as rainfall amounts, were observed at up to 36 sites, and then compared to the corresponding RADM grid-average predictions. Quantitative comparisons were performed on pairs of point observations and grid predictions using average fractional differences, defined as the differences in observations and predictions divided by the sum of the observations and predictions.

For OSCAR II and IV, it was found that predicted and observed sulfate and nitrate wet deposition generally agreed, to within a factor of 1.6. Good agreement was found between the regions of highest predicted and observed deposition and in areas where the rainfall amount was reasonably well predicted. The results for OSCAR I were, however, less conclusive and are subject to continued investigation. A factor thought to affect the accuracy of the model predictions is the degree to which the RADM grid-average rainfall patterns are representative of the point measurements.

Across all of the events, agreement between modeled and observed deposition within a factor of two or better was generally observed. The best agreement was found for sulfate deposition. The lack of accuracy of the precipitation predictions was found to be the major determinant of the discrepancies between model predictions and observations.

MAJOR DELIVERABLES

Deliverable	Date	Significance
Report on the sensitivity analysis of regional wet deposition to atmospheric processes and of nonlinearity in wet deposition.	1988	Provides an improved theoretical understanding of the importance of major chemical and cloud conditions using the RADM Scavenging Module.
Report on the Washington, DC mesoscale acidic deposition field study.	1988	Repeats the Philadelphia study for a city with less industrial nitrogen oxide emissions to 1) establish the influence of a city on nitrate deposition downwind of the city, and 2) test the hypothesis that the strong increases in nitrates result from mobile source emissions.
Report on the Western Atlantic Ocean Experiment (WATOX).	1988	Calculates a preliminary budget for the continent-ocean system and calculates air and chemical fluxes for flow off and along the Atlantic coast.
Report on the analysis of emissions change and source attribution using the RADM II and the Engineering Model.	1990	Provides an assessment of source attribution using the new models incorporating effects of nonlinearity.
Report on the analysis of nonlinearity based on the applications of RADM and the Engineering Model and preliminary results of the model evaluation field program.	1990	Provides an assessment of the importance of nonlinearity in the acidic deposition system, involving transport and transformation. Provides a reference against which to judge earlier estimates of the effect of emissions changes in a given source region on deposition to receptor areas.
Report on ANATEX 87 results.	1989	Provides a data base for testing the estimates from mathematical models of inert gas transport under more complex meteorological conditions.

Deliverable	Date	Significance
Report on the preliminary evaluation of RADM II against first 6 months of FY 1988 and FY 1989 field study data.	1990	Provides a first evaluation of the RADM II. Establishes the accuracy of using RADM for NAPAP assessment work.
Report on CURTAIN program boundary condition measurements for use by RADM.	1989	Provides an important data base to reduce major uncertainties in setting boundary conditions for the RADM model.
Report on methods and results for time trends and spatial patterns of wet deposition data.	1989	Provides a careful examination of trend analysis and points to problems associated with oversimplification in trend interpretations. Provides for more reliable trend estimates.
Report on the influence of meteorology and emissions on air quality and precipitation chemistry.	1989	Provides for a more informed separation of meteorological and emission factors in causing changes in concentrations and deposition fluxes. Important for the study of temporal trends.

ATMOSPHERIC DEPOSITION AND AIR QUALITY MONITORING

INTRODUCTION

The principal objective of the Task Group on Atmospheric Deposition and Air Quality Monitoring is to assure that consistent and uniform measurements of wet and dry deposition are available for long-term trend analysis, effects research, and other research. This requires the establishment and maintenance of high-quality nationwide monitoring programs which need to remain in operation for a period of decades.

In response to these needs, the Task Group has funded and helped design three monitoring networks: the National Trends Network (NTN), a fully deployed 150-station wet deposition network; the National Dry Deposition Network (NDDN), a partially implemented 75-station dry deposition and air quality monitoring network; and a smaller global background monitoring network, the Global Trends Network (GTN). The NTN design utilized the already existing National Atmospheric Deposition Program (NADP) network sponsored by the State Agricultural Experiment Stations. Since the NTN adopted NADP siting criteria and operating protocol, and shares the same analytical laboratory, the two networks merged and are known under the acronym NADP/NTN.

The NDDN has been designed and is being implemented in several phases in cooperation with the Atmospheric Modeling and Transport Task Group. The network is designed to provide regional rural air quality and meteorological data to be used for evaluating regional acid deposition models and for determining dry deposition and air quality trends in the eastern United States. The network will be expanded to the western United States in order to produce rural air quality and dry deposition data for use in conjunction with effects research. The present state of knowledge about the operation of dry deposition processes under complex terrain conditions will limit the number of sites at which dry deposition can be calculated during the next several years.

1990 RESEARCH OBJECTIVES

- To determine the spatial and temporal variations in the composition of atmospheric deposition within the United States by means of nationwide monitoring networks.
- To estimate the background composition of atmospheric deposition worldwide through the operation of deposition monitoring and research sites at remote locations (GTN).
- To provide air-quality measurements needed for the evaluation of atmospheric models and for assessment of effects due to atmospheric deposition.
- To develop methods for reliable determination of wet and dry atmospheric deposition.
- To support an integrated storage and retrieval system for atmospheric deposition and air quality data for the United States.

RESEARCH ACTIVITIES IN 1987

Wet Deposition Monitoring

The wet deposition monitoring program consists of the operation of 150 NTN wet deposition monitoring stations across the conterminous United States, supplemented with 50 NADP stations. Additional wet deposition data were obtained from other networks, such as those operated by several states and by the Electric Power Research Institute. In addition, 7 to 10 remote global sites were operated as part of the Global Trends Network (GTN). Monitoring stations are operated by the U.S. Geological Survey, National Park Service, Bureau of Land Management, U.S. Forest Service, National Oceanic and Atmospheric Administration, Environmental Protection Agency, U.S. Fish and Wildlife Service, Tennessee Valley Authority, State Agricultural Experiment Stations, Department of Energy National Laboratories, State agencies, universities, and private corporations. The samples are collected on a weekly basis according to a consistent and rigorous protocol. All activities from sampler function to final data compilation are subject to careful quality assurance and quality control procedures.

The NADP/NTN data, along with precipitation chemistry data from Canadian and private networks, are stored and compiled in the Acid Deposition System (ADS) data base on a yearly basis at the Pacific Northwest National Laboratory. The ADS data base also will include rain and cloud composition measurements from the Mountain Cloud Chemistry Network (funded by the Atmospheric Chemistry Task Group). Supplementary data from useful but less comprehensive networks are available to NAPAP users from this data base.

Where needed for intensive studies for model evaluation, parallel wet deposition samplers that operate on daily sampling schedules will be collocated at selected NADP/NTN stations. These samplers will be funded by other task groups but operated under protocols complementary to the NADP/NTN operations. These intensive studies will run periodically for about 2 years beginning in the summer of 1988.

Dry Deposition Monitoring

Close to emission sources, average dry deposition tends to be more important than wet deposition. At regional distances (hundreds of kilometers) in the eastern United States, dry deposition may be equal to or somewhat less than the wet component. The technology for measurement of dry deposition is under continuing development (see Chapter II, Atmospheric Chemistry), but enough is now known to justify the operation of an exploratory rural air quality network to estimate dry deposition. The network will measure airborne gaseous pollutants as well as particulates ($\leq 10\,\mu m$). As measurement techniques are refined by the Atmospheric Chemistry Task Group, and as measurements continue, the uncertainties in estimates of regional dry deposition will be successively reduced. A total of about 75 of these stations will be established by 1989 and then operated for the remainder of the NAPAP lifetime and possibly beyond. Air quality and meteorological data, as well as calculated dry deposition values (at selected sites), will be stored in an atmospheric deposition data base. As with wet deposition data, annual summaries will be produced, and measurements from other relevant stations will be included in the NAPAP air quality data base.

Wet Deposition Monitoring Research

This program supports wet deposition research at nine Multistate Atmospheric Power Production Pollution Study (MAP3S) event monitoring sites in the eastern United States, including expanded research at Whiteface Mountain, New York, and at Pennsylvania State University. (Drydeposition research is conducted under the Atmospheric Chemistry Task Group). At the MAP3S stations, samples are taken for each precipitation event and are sent for analysis to the Pacific Northwest Laboratory in Richland, Washington. This part of the research effort focuses on improvement of deposition collection accuracy, efficiency, and protocols for the routine monitoring networks and collection of data for source/receptor modeling.

A joint U.S.-Canadian comparison of the results of precipitation chemistry monitoring through the NADP/NTN and the Canadian Air and Precipitation Monitoring Network (CAPMoN) began in 1986. The program includes the use of both NADP/NTN and CAPMoN precipitation collection equipment and field protocols at sites at State College, Pennsylvania and Sutton, Quebec. For this comparison, NADP/NTN samples are analyzed at the network's central analytical laboratory at the Illinois State Water Survey, and the CAPMoN samples by Environment Canada's National Water Quality Laboratory. Each network provided and installed its equipment at both sites, trained the operators in the procedures of their respective networks, and performs laboratory and field quality assurance operations according to their respective protocols.

FUTURE RESEARCH ACTIVITY

The Task Group's plans for 1988 and 1989 include the continuation of existing monitoring networks, the implementation of the full NDDN, and the continuation of research activities at the MAP3S wet-deposition research stations.

The 75-site NDDN will be implemented by 1989, and data from these sites will be provided to assist in the model evaluation program under the Atmospheric Modeling and Transport Task Group. Research at several NADP/NTN sites will place increased emphasis on accurate measurement of the deposition of acidic pollutants in snow and effects associated with aging or melting of the snowpack.

1987 RESEARCH HIGHLIGHTS

- Annual data summaries of precipitation chemistry in the United States for 1985 and 1986 were published by the NADP/NTN Coordinator's Office at Colorado State University. The annual mean hydrogen ion concentrations for 1986 (1987 data are currently under analysis) are shown in Figure IV-1. Several organizations, including the U.S. Geological Survey, the Environmental Protection Agency, and the USDA Cooperative State Research Service are involved in analyzing and summarizing data from the NADP/NTN.

- Quality assurance data indicate that the field analysis of pH has improved for the NADP/NTN; over 80 percent of the measurements meet network quality control criteria. Based on interlaboratory comparison data, the laboratories for the CAPMoN, NADP/NTN, and Utility Acid Precipitation Study Program (UAPSP) are producing comparable results for major inorganic constituents in precipitation. However, other studies have shown that there is a neutralization of acidity occurring during the sample collection, field handling, and sample transportation within the NADP/NTN. The extent of the neutralization is from 6 to 10 microequivalents of hydrogen ion per liter. This is approximately equivalent to a change of only 0.1 pH unit at a pH of 4.3, but represents a larger underestimation of acidity at higher pH values. This process may explain the long-observed differences between field and laboratory pH measurements.

- An analysis of the NTN's siting design was performed to evaluate network sensitivity, site redundancy, and regional representativeness of sites. The network was found to be capable of detecting annual changes of 20 to 30 percent in hydrogen ion, sulfate, or nitrate concentration in the eastern United States in regions with dimensions of 4 degrees of longitude and latitude (approximately 160,000 km^2). The sites chosen for the network appear to be regionally representative, with only two sites showing any evidence of being influenced by local sources. There is no evidence of site redundancy.

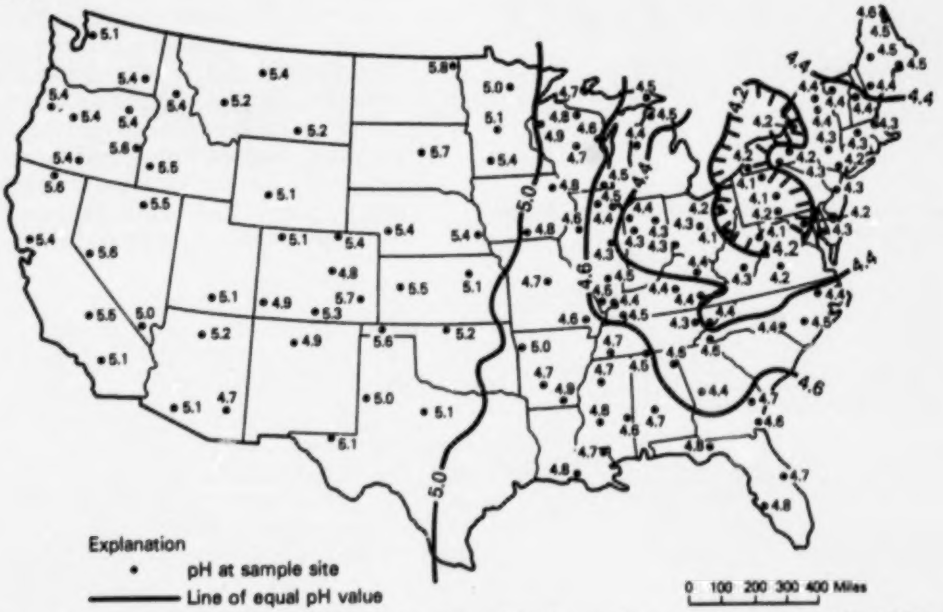

Figure IV-1. pH of wet deposition in 1986 (precipitation-weighted annual average) based on NADP/NTN data.

- A new cooperative station was established in the Global Trends Network (GTN), sponsored jointly by NOAA and EPA in collaboration with the People's Republic of China. The site is located in Yunnan Province at Lijiang in the southwest part of the People's Republic of China. Data from the first year of operation have been analyzed and indicate that the site is an excellent remote continental location. Sulfate concentration in precipitation is similar to that of the other long-term remote sites in the GTN. Results of the GTN are provided in Table IV-1.

The average acidity of precipitation at these stations ranges from a pH of 4.7 to 5.2. Because the GTN measures organic acids while the NTN does not, the GTN and NTN results are not directly comparable for pH. Allowing for this difference, however, the average pH in the eastern United States is lower than at remote sites throughout the world. Concentrations of ammonia, nitrate, and sulfate are much higher in the eastern United States than at remote sites.

- A site-location study for adding five air quality monitoring sites to the NDDN in the southeastern United States was completed (see Figure IV-2). The selection priorities were as follows: identifying patterns of sulfur dioxide and ozone for trend analysis, identifying ambient concentration patterns for forest effects research, aquatic effects, and model evaluation. The design of the study was based on historical surface data as well as model output. The final selection included sites in Arkansas, Mississippi, Alabama, Georgia, and northern Florida.

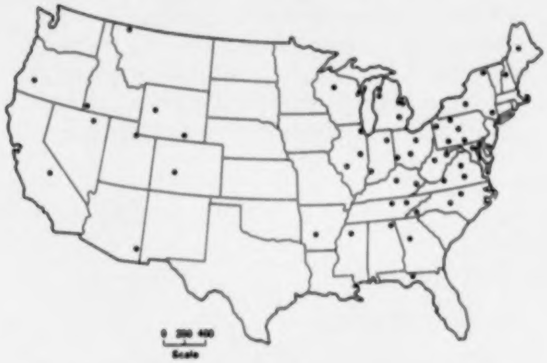

Figure IV-2. Location of dry deposition monitoring sites. Not all sites are implemented.

- The Multistate Atmospheric Power Production Pollution Study (MAP3S) completed its tenth year of operation. Current research at the Lewes, Delaware site continues to focus on the accurate measurement of trace elements (Al, Fe, Mn, Zn, Cu, Ni, V, Cd, Pb, As, and Se) in precipitation, and the evaluation of their utility as emission source tracers. For example, the concentrations of the coal combustion tracers arsenic and selenium exhibit strong correlations with the acid components in precipitation (hydrogen ion, nitrate, and excess sulfate).

- A precipitation chemistry sampling site using MAP3S protocol for event wet-only collections was established in a new research area in the Great Smoky Mountains National Park, North Carolina. The site is near Clingman's Dome at an elevation of 1800 m and is designed to test theories on differences in atmospheric deposition rates between high- and low-elevation forests. The initial year's data on wet deposition indicates the following: major ion concentrations at the high-elevation site in summer are lower than at the nearby, low-elevation Oak Ridge MAP3S site by factors of 1.5 to 1.9, while during the winter they are lower by factors of 1.6 to 3.2. During both seasons, the ratios of $SO_4^=/NO_3^-$ are comparable at both sites, while the ratio of $H^+/(SO_4^= + NO_3^-)$ is higher at the mountain site, indicating somewhat less acid neutralization at this site. Precipitation amounts are nearly twice as high at the mountain site, suggesting that the lower ion concentrations may be due to dilution, but that overall wet deposition to the two sites will be comparable.

Table IV-1. Global background precipitation chemistry compared to that of the Eastern United States.

Station	Katherine, Australia	Poker Flat, Alaska	Amstersdam Is., Indian Ocean	Cape Point, So. Africa	Eastern U.S. NTN Range [1]
Latitude:	14°S	67°N	38°S	33°S	
Longitude:	132°E	147°W	78°E	19°E	
Frequency of Collection:	Event	Event	Event	Event	Weekly
Period of Record:	Nov. 80 - Feb. 87	Nov. 79 - Sept. 82	May 80 - June 86	Aug. 83 - Sept. 86	1985
Volume weighted average:					
[2] pH:	4.72	4.95	5.08	5.24	4.2 to 4.6
			$\mu eq\ L^{-1}$		
[2] H^+	19.1	11.3	8.4	5.8	25 to 50
Ca^{2+}	2.1	1.4	11.4	19.9	4 to 8
Mg^{2+}	1.7	0.5	56.4	66.3	2 to 3
K^+	1.2	0.5	5.6	7.5	0.4 to 0.8
Na^+	4.9	0.9	253.	286.	2 to 8
NH_4^+	3.4	2.7	2.4	8.2	8 to 16
NO_3^-	4.5	2.4	1.6	3.9	15 to 30
Cl^-	8.5	3.0	298.	340.	4 to 8
SO_4^{2-}	4.2	9.1	[3]5.1	[3]6.6	35 to 65
[4] $HCOO_T$	11.9	4.3	2.8	2.2	---
[5] CH_3COO_T	4.6	1.2	1.1	0.5	---
[6] N	252	53	170	65	---
[7] N_{org}	201	14	129	65	---

Precipitation chemistry data from the Global Trends Network (GTN), a remote precipitation monitoring network, and from the eastern U.S. National Trends Network (NTN) sites. The purpose of the GTN network is to measure precipitation chemistry in remote locations around the world. The GTN is an event network which preserves organic acidity, unlike the NTN, which is a weekly network and does not preserve organic acids. Therefore, measurements of pH from the GTN and NTN are not directly comparable. The results are shown for the period of record. However, not all results are for the full period.

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1. The annual ranges presented here are the approximate ranges of pH and concentration as reported in the 1985 NADP/NTN Annual Data Summary. They reflect data for the area of the eastern United States north and east of a line drawn from Georgetown, South Carolina to Atlanta, Georgia to Erie, Pennsylvania. A few individual station observations in this region may be above or below these ranges. Further, the range shown for 1985 may not hold precisely for other years.

2. GTN and NTN pH and hydrogen ion concentrations are not directly comparable because organic acidity is not preserved in NTN samples.

3. Total sulfate concentration, i.e., sea salt and non-sea salt was 35.1 $\mu eq\ L^{-1}$ at Amsterdam Island, and 41.9 $\mu eq\ L^{-1}$ at Cape Point, South Africa.

4. $HCOO_T = HCOOH_{aq} + HCOO^-$.

5. $CH_3COO_T = CH_3COOH_{aq} + CH_3COO^-$.

6. The number of events sampled for inorganic ions at the GTN station; the NADP/NTN stations were sampled weekly. GTN stations with less than 50 events sampled and reported to date are not shown in this table.

7. The number of events sampled for formic and acetic acid at the GTN stations.

MAJOR DELIVERABLES

Deliverable	Date	Significance
1987 precipitation chemistry data for all NADP/NTN stations.	1988	Data will be available for interpretation by NAPAP scientists.
Summary reports and trends analysis of 1989 and earlier years' data.	1990	Will provide trend analysis for five years of data on full 150-site network.
Complete implementation of the NDDN.	1989	Will provide nationwide coverage of air quality and dry deposition monitoring.

TERRESTRIAL EFFECTS

Effects on Forests

INTRODUCTION

Terrestrial ecosystems in the United States have been subjected to elevated levels of atmospheric pollutants on a regional scale for several decades. In the Northeast, growth reductions are reported for red spruce at lower elevations. At some locations spruce-fir forests in the high elevations of the Appalachian Mountains appear to be experiencing higher than normal incidence of mortality. Other species, such as eastern white pine and sugar maple, also appear to be experiencing adverse changes. Forest inventory data in the Southeast suggest reduced growth over the past several decades for commercial pines. The reasons for the mortality and reduced growth in these areas are unknown. However, it is known that oxidants, such as ozone, cause visible damage to forests of the southern Sierra Nevada and the San Bernardino Mountains of California. The overall influence of pollutants on forest health in these areas as well as in other regions is being investigated in Task Group programs.

1990 RESEARCH OBJECTIVES

- Determine the effect on productivity and health of major trees from exposure of the foliage to acidic deposition, gaseous sulfur dioxide and nitrogen oxides, and associated oxidants in exposure-response studies (foliar-mediated effects).
- Estimate the long-term effect on productivity and forest health from acidic deposition on a wide variety of forest soils at near-ambient levels (soil-mediated effects).
- Identify the major factors causing visible damage to forests above cloudbase in the mountains of the eastern United States.
- Initiate biochemical, physiological, and ecological research to study whether causal relationships exist between acidic deposition, related air pollutants, and natural stresses (e.g., climate, pathogens) and the condition of forests where adverse changes are demonstrable (mid-term to long-term mechanistic studies).
- Establish a long-term program for monitoring forest condition.

1987 RESEARCH ACTIVITIES

The forestry research program is supported and managed jointly by the USDA Forest Service, the U.S. Environmental Protection Agency and the Tennessee Valley Authority, with additional support from the forest products industries. This Forest Response Program is focused in four forest regions: the spruce-fir forests of the eastern United States, the commercial pine forests of the Southeast, eastern hardwood forests, and the coniferous forests of the West.

Research in the northeastern spruce-fir forest is designed to test several hypotheses of forest decline including nutrient leaching from the foliage due to acidic fog and rain, effects of excess nitrogen (from acidic deposition) and ozone exposure on frosthardiness of spruce and fir, and soil changes due to acidic deposition. Southern conifer forests show an apparent growth decline in plantations that are not intensively managed. Atmospheric pollutant exposure may be a cause of this decline. Other possible causes such as drier conditions and increased hardwood competition are also being investigated. Highest research priority has been placed on tree physiological processes directly related to growth and on soil and soil microorganism processes which affect plant growth.

The eastern hardwoods and western conifers research programs were developed more recently and were established to define the extent of the forest decline problem, if any, before addressing the causes. Some controlled exposure studies are being conducted to help define the potential risk to selected species. The eastern hardwoods region is in an area of high atmospheric deposition and includes areas with several unexplained declines of tree species, including sugar maple. Research sites are being studied across air pollution gradients in these areas. The western conifers research program includes areas where oxidant damage is probably the most severe in the nation. An objective of the program is to determine those areas in the West where pollution-sensitive forest tree species occur in combination with severe atmospheric pollution.

FUTURE RESEARCH ACTIVITY

The 1988 research activities will represent a new phase in the forestry research program. In the first 2 to 3 years, research was initiated across a wide range of topics. Starting in 1988, the program will become more focused. Work for 1983 will build on the existing program, get maximum use from current projects, and add some new studies that are required to understand a complex situation. Priorities were established for new research to emphasize: a) work critical to the program's overall success, but not yet in place; b) research needed to understand and interpret work in progress or already completed; and c) work that can be adequately addressed during the latter part of the program and therefore contribute to the final NAPAP assessment.

A high priority was given to atmospheric exposure studies, seedling exposure studies, nutrient and carbohydrate status studies, modeling, and monitoring forest condition. Medium priority was given to competition studies, and lower priority to mixed species stand modeling and long-term soil dynamics.

1987 RESEARCH HIGHLIGHTS

- The growth of loblolly pine seedlings is reduced by ozone, and the magnitude of this response varies among families. Researchers with the Southern Commercial Forest Research Cooperative examined commercially preferred families of loblolly pine from throughout the southern commercial pine region for sensitivity to ozone. There were differences in sensitivity to ozone among the families studied, but generally the height, diameter, and total biomass decreased as ozone concentrations increased. These studies were done with 12-week-old seedlings exposed to five levels of ozone (0, 80, 160, 240 and 320 ppb) for 6 hours/day, 4 days/wk for 12 weeks. No tests were conducted within the ambient range (40 to 60 ppb for 7 hours/day, 7 days/wk) of ozone concentrations; however, open-top chamber studies are underway which will provide the data necessary for interpolation. Ongoing studies are attempting to determine possible cumulative impacts and effects from ozone/acidic rain combinations.

- At the current nitrogen deposition rate in the southeastern United States, nitrogen is not toxic to mycorrhizae. This conclusion was reached following an exhaustive literature review and evaluation of known nitrogen treatment levels in forest management practices. It is possible that mycorrhizae are affected indirectly through changes in tree nutrition and health. Preliminary findings in controlled exposure experiments have demonstrated a reduction in mycorrhizal infection of short roots and reduction of root starch following ozone treatments (160 ppb) in the field, but not in greenhouse experiments.

- The spruce-fir forest at high elevations in the eastern United States show severe damage in some areas. Spruce mortality at Whiteface Mountain, New York averaged 25 percent below 1,000 m and 60 to 65 percent above 1,000 m. Mortality is greatest on northwest facing slopes at 1,000 m, where 75 percent of trees and 80 percent of stem basal area were dead. Pollutant concentrations in air and cloud water at these high elevations are one of the possible causes of the decline. Although daytime average ozone concentrations are similar to those at low elevations, nighttime concentrations sometimes remain elevated on these mountaintops compared to the diurnal variations observed at nearby lower-elevation sites. Cloudwater pH averaged 3.7 on Whiteface Mountain during May-October 1986, and exposure of trees to cloudwater is much more frequent at high elevations than at low elevations.

- All low-elevation New England forest tree species investigated are growing at rates equal to or exceeding those prior to 1960, with the exception of red spruce and balsam fir. Recent studies have identified a definite decline in basal area growth rates of stands (60 to 75 years old) of red spruce and balsam fir starting about 1960. It once was suspected that the decline was associated with acid rain. However, a recent analysis of tree ring data indicates that stand aging is playing a major role in these growth declines in low-elevation forests. These data showed that most low-elevation red spruce stands are functionally even-aged as the result of harvesting and spruce budworm infestations after the turn of the century. These stands have now attained an age when a natural reduction in growth rate is expected.

- A manual of symptoms of damage on Eastern forest trees is now available. This manual depicts symptoms recognized as resulting from atmospheric deposition and differentiates them from symptoms from other causes that might be erroneously attributed to atmospheric deposition. The manual is available as a guide to help in the characterization of different types of forest damage.

MAJOR DELIVERABLES

Deliverable	Date	Significance
Evaluate tree studies of low-elevation red spruce.	1988	Contributes to a decision to continue or terminate research on low-elevation spruce forests and concentrate efforts on high-elevation forests.
Quantify changes in forest condition across a pollutant gradient in Pennsylvania.	1988	Assesses growth in hardwood stands across a pollutant gradient and defines the spatial context of any growth response for a large geographic area (i.e., Pennsylvania).
Quantify seedling response to sulfur, nitrogen and associated pollutants under ambient conditions.	1988	Summary of available dose/response data for seedlings of major tree species. Will provide information on the role of airborne pollutants in forest health.
Progress report on the analysis analysis of growth trends in southern pine plantations.	1988	Report confirms declines in stand basal area growth for naturally regenerated loblolly, shortleaf and slash pine stands that are unexplained by stand dynamics.
Assess impact of sulfate and nitrogen deposition of southern forest soils.	1988	Review dealing with soil response to acidic deposition will discuss whether Southern soils are or will be susceptible to chemical change.
Develop rationale and design for forest health monitoring.	1988	Permanent forest health survey plots will be recommended for testing and implementation in the 1988-89 field season.
Evaluate the extent and magnitude of recent changes in forests.	1988	Provides current description of U.S. forest condition and discusses whether changes are in progress for which causes are uncertain.

Effects on Agricultural Crops

INTRODUCTION

Because most research studies have indicated that ambient levels of acidic rain alone do not appear to reduce crop yield directly, current research efforts focus on possible indirect effects by examining the interaction of acidic rain with other stress factors such as gaseous pollutants, disease organisms, and drought. Studies to examine the mechanisms of ozone damage to crops are being continued because it is known that this pollutant causes significant effects at ambient concentrations. The agricultural crops research program is supported by the USDA Cooperative State Research Service, the Agricultural Research Service and other USDA sources.

The goals of the 1987 crops research program were the following:

- Establish exposure-response relationships for acidic deposition and related gaseous pollutants with the yield of both nationally and regionally important agricultural crops.

- Develop information on the mechanisms by which air pollution damage occurs in sensitive crop systems to support exposure-response models and to justify extrapolations.

- Provide data on interactions that will contribute to the information needed to assess effects on a regional scale by determining whether climate, soil type, biotic stresses (such as insects), and other air pollutants influence plant response to acidic deposition.

1987 RESEARCH ACTIVITIES

Exposure-Response

The results from field investigations have indicated that under typical growing conditions, there are few differences in the response of the major crop plants (soybean, corn, wheat) exposed to simulated rain with high ambient levels of acidity (pH 3.5-4.5) compared to simulated rain with low levels of acidity (pH $\geq$ 5.0). Recent investigations with other crop species such as oat, potato and sorghum have supported the conclusion that acidic rain is not an important direct influence on crop production.

Screening studies in which greenhouse-grown crop seedlings are exposed to high levels of acidity in rain have indicated that crop species vary in their sensitivity to the treatments. Relative foliar injury from a one-hour rain event at pH 2.5 may predict growth effects in seedlings exposed to 15 one-hour rain events at pH 3.0. However, it is not known whether this type of screening procedure can adequately predict potential yield effects on field-grown crops from ambient levels of rain acidity.

Greenhouse experiments were initiated to determine whether effects of simulated acidic rain (pH levels of 4.9, 4.2, 3.5, and 2.8) on rhizosphere microorganisms of a sorghum x Sudan grass hybrid were plant- or soil-mediated. Preliminary results indicated increased root and shoot biomass and increased population densities of several types of bacteria and fungi as the acidity of the simulated rain increased. Most effects were apparently soil-mediated because a response was not observed when the soil was covered and only the foliage was sprayed with simulated rain.

A unique exposure situation exists in some regions of California where locally important crops are sometimes exposed to high levels of ozone and also to fogs with higher levels of acidity (average pH $\sim$ 3.0) than those that occur in rain. In a study using simulated fog with pH levels of 7.24, 2.69 and 1.68, the yields of strawberry, tomato, pepper and alfalfa were reduced only by repeated applications of pH 1.68 fog compared to pH 7.24 fog. Celery was unaffected by any of the acidic fog treatments. In contrast, all of the crops exhibited biomass decreases from ambient levels of ozone (12-hour seasonal average ozone = $129\,\mu g\ m^{-3}$/64 ppb), except for strawberry. The combined effects of pH 1.68 fog and ambient ozone were additive in all five crops examined.

Previous work, which indicated that the yield of most tested crops are reduced by exposure to ozone concentrations typical of the region in question, was expanded with the testing of additional crops. Results from studies in the Midwest and Southeast indicate that both muskmelon and watermelon are sensitive to ozone at levels within the ambient range, although cultivars varied in their sensitivity.

Mechanisms

Several studies have demonstrated that stage of development is an important factor affecting plant response to highly acidic rain. Reductions in the numbers of female flowers of cucumber plants and reduced pollen germination in corn occurred as a result of treatment with acidic rain, but these observations were not manifested in an effect on crop yield under normal growing conditions. However, studies with corn in which the quantity of pollen was severely limited by moisture stress resulted in reduced numbers of fertilized kernels per ear in the plants treated with pH 3.6 rain compared to pH 5.6 rain. Another study reported that a single one-hour acidic rain treatment reduced the percentage of corn kernels produced in plants when the rain treatment followed artificial pollination by one hour but not when the rain treatment ended ten minutes before pollination. These results suggest that an acidic (i.e., in the high ambient range) rain event that occurs during pollination may reduce corn yield when other stress conditions have limited the amount of pollen produced or the length of time for pollination. Studies with wheat and cotton did not demonstrate an effect of highly acidic rain on pollination.

Ethylene release from plant foliage has been correlated with the level of several plant stresses, including ozone and sulfur dioxide, and thus has been used as a diagnostic tool for determining pollutant sensitivity. However, no direct relationship was observed between ethylene production and rain treatment acidity (pH 5.6, 4.6, 3.8, and 2.8) in potato, radish, soybean and oat. Exposure of oat plants to 30 ppb ozone induced ethylene emission, but addition of acidic rain treatments did not change the level of ethylene emission. These results support those of the ozone-acidic fog studies which also reported no interactions.

A number of studies have indicated that ozone exposure can accelerate senescence (aging) in plants. The mechanism for this effect is being examined and studies with potato have indicated that ozone reduces the activity of a key plant enzyme, rubisco, by stimulating the degradation of this protein. It is not known whether this is a direct effect. Studies are continuing to determine whether ozone exposures that simulate weather-related inversions during the plant's life cycle, influence the quantity and activity of this enzyme as a direct result of the inversion or as a result of cumulative damage that is not observed until senescence.

Several chemicals have been identified which protect plants from pollutant damage. Paclobutrazol (PBZ), a growth retardant, protects plants from SO_2 injury. Ethylenediurea (EDU) is known to protect some plants from ozone injury. Studies are being conducted to determine how PBZ and EDU prevent ozone injury with the expectation that this may provide information on the mechanisms of ozone action on plants.

Ozone exposure (0, 5, 10, and 15 ppb, 6 hours/day, 5 days/wk for 8 weeks) of a pasture crop (subterranean clover) suggested that ozone induces an alteration of the carbon relations among the clover and its two microbial symbionts. The ozone suppressed dry weight of the plant root and the proportion of shoot nitrogen that had been fixed from the air. These results indicate that ozone can affect plant carbon and nutrient relations through effects on associated microbes.

Interactions

The results discussed in the previous section for corn pollen may also explain the mechanism responsible for an interactive effect on yield observed for corn plants exposed to acidic rain and drought. Further studies are needed to examine this possible mechanism of acidic rain damage because an acidic rain-drought interaction has been observed only in one of two cultivars, at one of two research sites and in only one of two years at the site where the effect was observed. Results of field studies with soybeans indicate that the effects of ozone on crop yield may be reduced by drought stress. High relative humidity during ozone exposure, however appears to increase ozone damage, as demonstrated in experiments with geranium in which reduced seed set occurred after a 6-hour exposure to 0.30 ppm ozone at 90 percent relative humidity but not at low humidity.

Combinations of the acidic rain precursors, sulfur dioxide (0, 10, 20, 30, and 40 ppb) and nitrogen dioxide (0, 40 ppb) for 4 hrs/day, 5 days/wk for four weeks, were reported to have synergistic effects by reducing growth of greenhouse-grown tomato plants more than either pollutant alone. Tomato growth at intermediate SO_2 levels was greater than at lower or higher levels, regardless of calcium levels in the potting mix.

Several studies were completed to examine the interactive effects on crops of insect and disease organisms with pollutants. Studies with soybean indicated that defoliation by Mexican bean beetle increased significantly with increasing ozone concentration and that larvae tended to weigh more and develop faster when reared on foliage exposed to ozone.

Laboratory and greenhouse studies indicated that highly acidic ($pH < 3.2$) rain reduced the severity of late blight disease of potato; however, this effect was not observed in field studies using pH 2.8 rain when exposure duration was lower. It is unlikely that ambient levels of acidic rain will prevent late blight disease, although these results suggest that the disease effects are not exacerbated by acidic rain.

In inoculation experiments, bacterial infections causing damage to field-grown lettuce and celery were increased by fog exposures at acidity levels $< pH 2.0$ compared to plants not treated with fog exposures. Spinach plants treated with pH 2.4 and 2.0 fog had more injury from cabbage loopers but less injury from an aphid-transmitted virus. Other studies have indicated that simulated acidic rain may accelerate the washoff of some fungicides. These experiments indicate that the indirect effects of highly acidic fog may influence the cost to growers for pest control and change pest management strategies.

FUTURE RESEARCH ACTIVITY

Research activities in 1988 will address the following specific objectives:

1. Evaluate the influence of exposure dynamics on crop response to acidic rain, gaseous pollutants, and aerosols or fog.

2. Conduct further investigations to examine the interaction of acidic wet deposition, dry deposition and drought on corn yield. Effects of acidic precipitation on soil-inhabiting invertebrates in corn systems will also be examined.

3. Determine potential interactions of ozone and acidic rain by examining growth, yield and disease development in soybean exposed to these pollutants singly and in combination.

4. Assess the mechanisms of ozone and acidic rain damage to crop plants through biochemical and physiological studies using tissue cultures and whole plants.

5. Evaluate the effects of acidic rain, including sulfur:nitrogen ratios, on the chemistry of the soil surrounding the root systems (rhizosphere) of various crop species.

6. Examine the direct effects of simulated acidic rain on the sexual reproduction of corn in greater detail to determine possible mechanisms for the effect and its significance on yield.

7. Determine the effects of ozone and sulfur dioxide on three watermelon cultivars with differing sensitivities to ozone.

8. Test the degree of ozone resistance in clover isolates that have been identified and separated from a field exposure experiment for eventual studies of the genetic basis for ozone tolerance.

1987 RESEARCH HIGHLIGHTS

- Recent investigations have supported the conclusion based on past studies that ambient levels of acidic rain have little or no significant direct effects on crop yield under typical field conditions. This is in contrast to the damaging effects on crop yield observed for ozone at ambient concentrations.

- Current studies which focus on more complex interactions have indicated that indirect and more subtle effects of acidic rain and ozone may affect crop production by affecting the severity of damage caused by drought, insect infestation, and disease organisms. Generally, these effects have been observed only at higher than ambient levels of acidity.

- In the unique exposure situation that exists in some areas of California where levels of ozone and fog acidity are extremely high, there is a greater likelihood of damage from these pollutants than in other regions of the United States. Research results, however, do not demonstrate interactive effects of ozone and acidic fog. This result supports the lack of interaction observed for ozone and acidic rain in other studies.

MAJOR DELIVERABLES

Deliverable	Date	Significance
Report on the influence of ozone and acidic deposition on rhizosphere microorganisms of sorghum.	1988	Gives needed information on the effects of pollutants on below-ground organisms associated with a crop plant.
Publication describing the measurement of dry deposition of $SO_4^{=}$ and NO_3^{-} to soybeans.	1988	Describes the importance of dry deposition to the leaf surface microenvironment.
Publication describing the results of a screening study to test crop species and cultivars for relative sensitivity to simulated acidic rain.	1988	Gives information on possible techniques which can be used to quickly evaluate crop sensitivity to acidic rain and provides additional response data for crop species and cultivars not previously studied.
Report on the effects of simulated acidic rain on sexual reproduction of corn.	1988	Demonstrate the importance of plant reproductive stage and timing of acidic rain events.
Report on the effects of an antioxidant used in combination with acid rain treatments on an ozone-sensitive crop.	1988	Will aid in determining whether antioxidants are a viable research tool to use in studying the effects of ozone and acidic rain combinations.
Report on the results of field investigations using radiotracers to study the effects of ozone.	1988	Will provide information on the effect of ozone on carbon partitioning and nitrogen accumulation.

AQUATIC EFFECTS

INTRODUCTION

The research program for the Aquatic Effects Task Group is designed to quantify and characterize the subpopulation of surface waters at risk in the United States as a result of acidic deposition in terms of both chemical and biological effects. Forecasting the number of systems that might change in response to various deposition levels, and the rate and magnitude of change, is of particular interest. Regional surveys of water chemistry and watershed characteristics have served as the foundation for quantifying the present status of surface waters in potentially sensitive areas of the United States. Site-specific studies focusing on mechanisms of acidification have provided a better understanding of the processes that control surface water sensitivity to acidic deposition. By integrating both approaches into one research strategy, the characteristics of systems that are potentially sensitive to chemical and biological change can be identified. This characterization, in turn, leads to a projection of the types, locations, and proportion of systems that will be most likely to change as a result of acidic deposition. The Aquatic Effects program also focuses on the ecological effects of surface water mitigation (liming), and the degree to which changes in the quality of ground, cistern, and surface waters potentially affect human health.

1990 RESEARCH OBJECTIVES

- Quantify the extent, location, and characteristics of sensitive and acidic lakes and streams in the United States.
- Identify the factors that control sensitivity of surface waters to acidic deposition and quantify their relative importance.
- Project and verify the rate of change in surface water chemical composition as a function of acidic deposition.
- Determine the relationship between surface water acidification and stress on populations of freshwater organisms.
- Investigate the relationship between the chemistry of surface, cistern, and ground waters and human health.
- Evaluate the ecological effects of techniques used in a) restoring surface waters that are acidic, or b) protecting those waters that may acidify.

RESEARCH ACTIVITIES IN 1987

Chemical Status of Surface Waters

The present chemical status of surface waters is being examined through two major programs: Chronic Acidification and Short-Term (acute) Acidification. Chronic acidification is addressed primarily through the National Surface Water Survey (NSWS), and short-term acidification through the Episodic Response Project (ERP) and other site-specific episode studies. Surveys conducted from 1984 to 1986 provide the foundation for classifying surface waters with regard to their potential sensitivity to change, allowing particular subsets of lakes and streams to be selected for further analysis. The ERP is designed to address the magnitude, duration, frequency, and regional extent of low-pH/acid-neutralizing capacity (ANC) episodes that accompany storms and snowmelt. The episodes studies will involve intensive (site-specific) chemical and biological monitoring, as well as the development of empirical models for extrapolating results to the regional scale. In 1987, data sets from previous studies of episodic events were synthesized and used to provide preliminary regional-scale estimates of episodic acidification in regions of the eastern United States.

Factors Controlling Sensitivity

Three major programs focus on factors controlling surface water response to acidic deposition. The Watershed Studies Program includes site-specific, process-oriented research on aquatic ecosystems. The Watershed Process/Soil Surveys Program is investigating biogeochemical processes important in controlling the response of surface water chemistry to acidic deposition. Studies include hydrogen ion, sulfate ion, nitrate ion, and aluminum ion mobility in soils, base cation supply and mineral weathering, hydrologic pathways, and the role of organics in surface water acidification. In the Watershed/Surface Water Acidification Program, a watershed/stream system and a lake are being artificially acidified to examine the direct chemical and biological effects of acidification.

Projecting and Verifying Rates of Chemical Change

The Direct/Delayed Response Project (DDRP) is the primary program in which forecasts of changes in surface water chemistry are being made. Verifying the findings of other program areas and providing the means to detect subtle, but biologically relevant, changes in surface water chemistry are key components of long-term monitoring. This program is being designed to collect long-term data records of lakes and streams that can be extrapolated to regional populations of surface waters.

Biological Effects

Biological effects studies are defining threshold concentrations of chemicals in surface waters that can adversely affect biota. Physiological and population-level effects from both chronic and episodic exposures are being examined. The focus is on key fish species, with secondary emphasis on primary producers, zooplankton, benthic invertebrates, amphibians, and waterfowl. Principal activities include defining the general types of biological change that result from acidic deposition and changes in water chemistry, identifying chemical conditions associated with biological effects, quantifying the relationship between the population status of selected fish species and changes in water chemistry associated with acidic deposition, and applying models on a regional scale to estimate biological effects associated with selected deposition scenarios through predictions of change in surface water chemistry.

Indirect Human Health Effects

The Indirect Human Health program area is evaluating the risk posed to human health by acidic deposition through two programs: 1) Effects of Acidic Deposition on Drinking Water and 2) Bioaccumulation of Metals. Drinking water studies include examining changes in water quality in cisterns and shallow water aquifers, as well as determining the potential risk posed to the population relying on unregulated or untreated drinking water sources. Bioaccumulation studies focus on metals that are potentially toxic to humans, particularly those that are concentrated in fish tissues. Currently, the regional distribution of mercury in fish tissues is being examined in the Upper Midwest. Mercury dynamics and cycling in lakes are also being investigated. An examination of the concentrations and mobilization of metals in sediments of waters with low ANC/pH is part of this program element.

Mitigation

Examining the ecological effects of liming as a mitigative option is the central focus of the mitigation program area. Six projects are evaluating the efficacy of liming for restoring acidic surface waters, including the most efficient means of application, the effects on surface water chemistry, and the effects on biota. The five-year mitigation research program on streams in the Northeast, Mid-Atlantic, and Southeast, and on a lake in the Upper Midwest is in the final phase of pretreatment data collection for baseline monitoring prior to mitigative liming. The physical, chemical, and biological baseline data are being used to determine the specific mitigative treatment required for each site and to provide a basis for determining the effects of treatment.

FUTURE RESEARCH ACTIVITY

Research in the Chronic Acidification Program will shift from data collection to data analysis in order to refine resource estimates and develop classification criteria for sensitive systems. The Phase II NSWS results on seasonal and spatial variability in lake chemistry will help refine the conclusions on current chemical status based on the single fall index sample collected in Phase I of the NSWS. The National Stream Survey (NSS) results will provide information on the present chemical status of streams in the Mid-Atlantic and Southeast. The NSWS and other data bases are being examined to investigate various acidification hypotheses, e.g., the importance of organics in acidification and the regional relationships between sulfate deposition and current surface water chemistry.

Examining acute acidification associated with deposition and natural factors will be the primary field effort in 1988 and beyond. Using index chemistry, model-based predictions will be made of how current resource status changes when episodes are considered. By 1990, these studies will be used to make preliminary regional estimates of the magnitude, duration, and frequency of episodes in eastern streams. A major effort to quantify the effects of episodic chemical changes on biota will be implemented as part of the ERP. These episodes studies will include elements that (1) determine whether episodic chemical changes in streams have long-term impacts on fish populations and (2) evaluate the processes and factors that influence the severity of episodic chemical changes and their associated effects on fish.

In 1988, the numbers and proportions of surface waters expected to become chronically acidic under current levels of deposition will be forecast for the Northeast and Southern Blue Ridge Province. Relationships between regional patterns of watershed characteristics (e.g., soils attributes, vegetation, and hydrology) and water chemistry for these two regions will be established and uncertainty analyses conducted. A third regional study will be initiated by the DDRP in the Mid-Appalachians, an area believed to represent a transition zone between the Northeast and Southeast in terms of soils, sulfate retention characteristics and other factors.

Watershed manipulation and site-specific studies will continue to provide data to evaluate acidification models, including information on hydrologic routing, sulfur cycling, base cation resupply, and aluminum mobilization. The conceptual design and research plan for a study that will increase confidence in forecasts concerning acidification in the Southeast will be completed. Research efforts related to the recovery of acidified systems in response to decreases in acidic deposition also will be expanded in 1988.

Biological effects research will focus on mercury and episodic chemical changes. Results of a fisheries survey in the Upper Peninsula of Michigan will be used to evaluate the relationships among fish presence, tissue mercury concentrations, and water chemistry. The development of a model to evaluate chronic acidification effects on fish in the Adirondacks and the Upper Midwest will continue.

Plans to initiate a regionally integrated long-term monitoring project including both lakes and streams will be finalized in 1989. Implementation is scheduled for FY 1989.

Indirect human health effects research will shift emphasis from evaluation of rainwater cistern drinking water supplies to shallow aquifers. Evaluation of existing groundwater data will be emphasized during 1988. Data collected during the regional survey of mercury bioaccumulation in fish tissue will be evaluated.

In the spring of 1988, limestone treatments will begin at the four mitigation research sites. Water quality changes and biological responses will be monitored closely during the post-treatment phase to determine the effectiveness and ecological consequences of the treatment.

1987 RESEARCH HIGHLIGHTS

Chemical Status of Surface Waters

Chronic Acidification: Data from the Eastern Lake Survey - Phase I (ELS-I) were used to determine chemical conditions in acidic and nonacidic lakes. Acidic lakes (ANC < 0) in the Northeast and Upper Midwest had similar median concentrations of sulfate but lower median concentrations of dissolved organic carbon (DOC) than did nonacidic lakes. Organic anions (calculated from DOC and pH) may contribute to the acidity of some acidic lakes in the Northeast in varying degrees, but their low concentration on a regional scale suggests that most acidic lakes in this region are currently acidic because of mineral, and not organic, acidity. As in parts of the Northeast, sulfate was the dominant anion in most acidic lakes in the Upper Midwest. This description of current patterns of lake acidity may not reflect the process by which the lakes became acidic. It is not clear to what extent some of the lakes originally may have been acidic because of higher organic anion and lower base cation content. Organic acidity in some Upper Midwestern lakes appears to be more important than was observed in the Northeast. In the Southern Blue Ridge, no acidic lakes or reserviors were observed. In Florida, the acidity of the Okefenokee is attributable to natural organic acids. The origin of acidity in the Florida Panhandle is undetermined.

In another analysis of ELS-I data, median lake sulfate concentrations in lakes between 4 and 2000 hectares were found to be highly correlated with estimated wet sulfate deposition in most NSWS regions (Figure VI-1). A positive relationship between lake sulfate concentration and calcium and magnesium concentrations is evident for the full northeastern data base. This reflects the fact that one way in which watersheds respond to acidic inputs is by releasing base cations such as calcium and magnesium. While this relationship between calcium and magnesium and sulfate applies to the northeastern sample, it may not apply to every individual lake or some subpopulation of lakes. In some systems, the acidic input may be completely offset by a release of calcium and magnesium with no resultant change in ANC as a result of acidic deposition. In other systems, however, the acidic input may be only partially offset by watershed releases of base cations. In these instances, acidic deposition would reduce ANC. These varying processes may explain the lack of a relationship between lake water sulfate and ANC in the Northeast. Detailed understanding of the relationship between the deposition of both basic and acidic substances and changes in lake chemistry is a key part of the Aquatic Effects Research Program.

An evaluation of low-conductivity lakes with low dissolved organic carbon sampled during ELS-I was recently completed in the Upper Midwest. A west-to-east increasing gradient in sulfate deposition occurs in this region. Data from regional lake surveys (Table VI-1) and the National Atmospheric Deposition Program indicate that the frequency and spatial distribution of acidic lakes in this region is correlated with the hydrogen ion concentration in atmospheric deposition, supporting the hypothesis that the deposition of hydrogen ions causes acidification of some lakes in areas of high deposition.

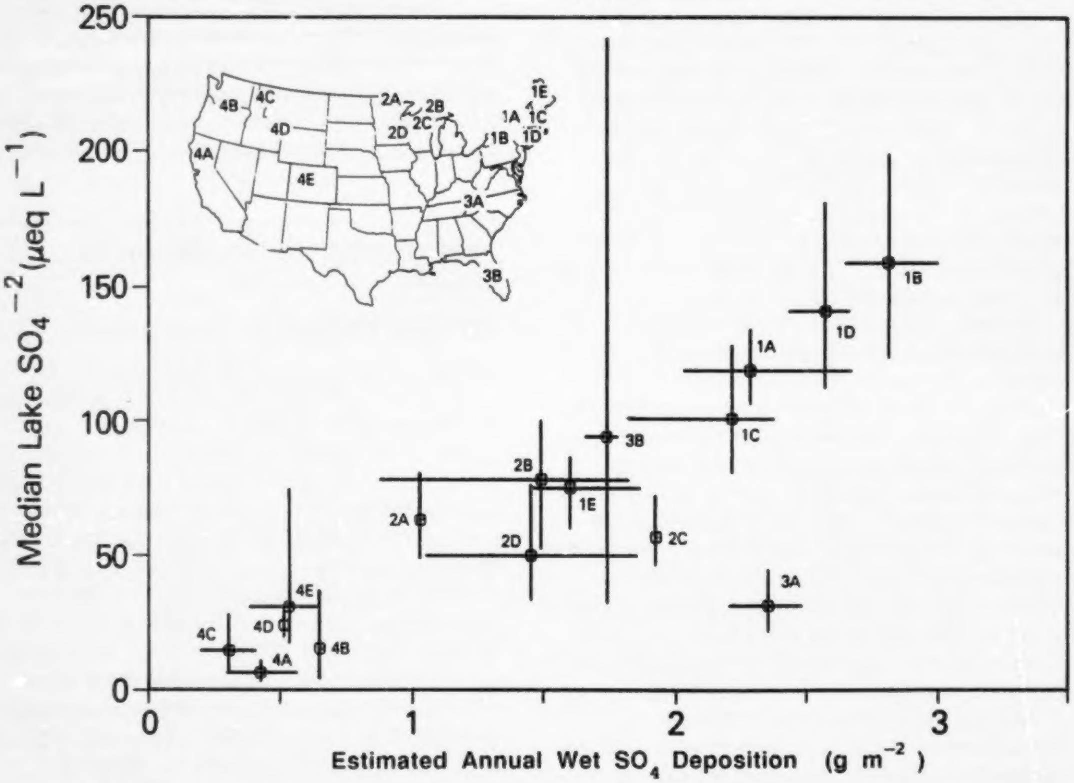

Figure VI-1. Median (circles) and 25th and 75th percentile (bars) of measured lake sulfate concentration and estimated sulfate deposition for subregions of the National Lake Survey. (Data from Sullivan et al., in press).

TABLE VI-1. Frequency of Occurrence of Acidic Lakes in Three Potentially Sensitive Areas of the Upper Midwest, Relative to the pH of Wet Deposition

Source of Data	Number of Lakes Studied	Criterion Used To Define Acidic Lakes	Northeastern Minnesota	Northcentral Wisconsin	Eastern Peninsula
			(percent of lakes studied)		
Nichols and Verry (1985)[a]	479	pH < 5.3	0	10	10
Schnoor et al. (1986)	772	ANC < 0	0	4	19
Glass et al. (1986)	861	ANC < 0	0	4	13
Linthurst et al. (1986)[b]	451	ANC < 0	0	3	18
NADP, deposition pH[c]			4.9	4.6-4.7	4.3-4.4

a. Information provided is for lakes in three national forests: Superior (MN), Nicolet (WI), and Hiawatha (MI).
b. Percentages shown are weighted population estimates.
c. NADP = National Atmospheric Deposition Program, volume-weighted mean pH of wet deposition (from Glass and Loucks, 1986).

A comparison of recent and historical chemistry data for Northcentral Wisconsin lakes and paleolimnological analysis of sediment diatom remains in an acidic Michigan lake, however, provided little or no support for this hypothesis. A recent analysis of deposition data indicates that in the Midwest a high percentage of the sulfate anions are associated with cations other than hydrogen. Further work is proposed to critically test the hypothesis that lake acidification is caused by acidic deposition in the Upper Midwest and to determine the source of the hydrogen ions.

The utility of cesium isotope Cs-137 as a dating technique in paleolimnological studies of Adirondack lakes was evaluated. Dating of sediments with Cs-137 to provide a continuous record of past lake and watershed changes is imprecise in Adirondack lakes having low levels of binding clay minerals and high levels of organic materials. Other work is being conducted on dating sediments using the natural isotope Pb-210. The work to date suggests that Pb-210 data are useful, but that careful interpretation is required.

Episodic Acidification: Decreases in ANC and pH during rainstorm and snowmelt events were observed in six streams located in mountainous areas of New York, North Carolina, Pennsylvania, Tennessee, and Arkansas. Depressions of 0.5 to 1.3 pH units were measured during the majority of events sampled, confirming that short-term acidification during storm events is potentially a significant chemical phenomenon in some U.S. streams. The role of acidic deposition in short-term acidification is being investigated.

A two-box mixing model, based on the theory that streamflow can be chemically partitioned into "old" and "new" water, adequately predicts the minimum episodic ANC measured in six streams in the eastern United States. In addition, data from a group of lakes sampled during the ELS-I show that the fall ANC value can be used to predict the minimum episodic ANC measured at the outlets of these lakes during spring snowmelt. These results support the use of a model-based approach to estimating the regional extent of acidic episodes in lake and stream populations such as those sampled by the NSWS. Typical results of applying the two-box mixing model to the NSS data bases show that the proportion of acidic stream reaches (ANC ≤ 0 $\mu eq\ L^{-1}$) is two to five times higher in most subregions when episodes are considered. In several subregions where no acidic reaches were sampled at spring baseflow, 2 to 3 percent of the reaches were predicted to become acidic during the most severe episodes. This simplified representation of stream response has not yet been verified, nor does it provide any indication of the duration, frequency of occurrence, or biological significance of episodes.

Although not yet verified, an empirical model of lake episodes using Eastern Lake Survey data for the Adirondacks predicts that more than 35 percent of the lake outlets in that region are acidic during snowmelt, compared to an estimated 11 percent during the fall sampling period. Although these results suggest a potentially large underestimate in quantifying acidification in this region on the basis of fall chemistry alone, the biological significance of these projections is unclear. The chemistry of outlet waters during periods when lakes are stratified (as in spring) does not necessarily reflect the chemistry of the whole lake, and fish refugia are thus expected to be present during such events. For example, acid pulses resulting from snowmelt were shown to have profound effects on the chemistry of streams flowing into clearwater acidic Cone Pond in New Hampshire. Effects were limited to the pond's surface waters, however, suggesting that deeper waters could provide refuge for pond organisms and perhaps for freely mobile stream organisms.

In each of six clearwater northern Wisconsin lakes (mean pH 5.9 to 7.8), pH values annually range from 1.0 to 1.5 units, with the lowest values usually occurring in late winter and early spring. Detailed investigations in one lake indicate that seasonal pH depressions are caused largely by increases in carbon dioxide under the ice. Furthermore, despite an acidic snowpack, the pH depression in the lake could not be attributed to input of acidic meltwater because total alkalinity did not change. The biological consequences of this naturally induced pH depression, similar to those reported to induce biotic effects in other systems, are unclear.

Factors Controlling Surface Water Response

Site-specific studies are continuing to examine the relative importance of hydrologic patterns and chemically and biologically mediated processes in determining the flux of water and chemicals through watersheds. Accumulated snowpack can be a major source of the total ionic input to some ecosystems, but the entry of ions to streams during snowmelt depends on the melting pattern and flow path through soils. In the Isle Royale ecosystem, flow through surface soils rather than subsurface soils appears to be a major factor in determining the quality of water entering a stream. In the Loch Vale watershed in the Rocky Mountains, the replacement of lake water with snowmelt accounts for changes in water chemistry, including a temporary drop in pH and alkalinity. As the snow does not appear to be greatly influenced by acidic deposition, this is probably a natural, recurring phenomenon.

Biological uptake and transformation are important processes that affect the flow of ions through watersheds during ice-free months. Conversion of sulfate to organic forms in Loch Vale may be important in reducing sulfate transport to receiving waters. Microbial organisms in forest floor soils in the Coweeta Watershed in North Carolina rapidly converted sulfate to organic forms. Such biologically mediated pathways in the sulfur cycle might provide a buffering mechanism against potential acidic deposition effects by reducing mobility of sulfate, which in turn could reduce cation leaching. Studies conducted in Isle Royale and the Upper Peninsula of Michigan indicate that input sulfate rapidly enters surface waters in watersheds with glaciated soils. In time, it is possible that continued acidic deposition to these soils might deplete the soil buffering capacity (i.e., the exchangeable base cation capacity) and reduce the ANC in the surface waters.

The flux of nitrogen compounds through watersheds is also controlled to a considerable degree by biological processes. In a study conducted in the west-central Adirondacks during snowmelt, the natural process of nitrogen mineralization, followed by nitrification (conversion to nitrate) in the forest floor, may be important in contributing to elevated hydrogen ion and nitrate levels in streams during snowmelt. Biological uptake and transformation of nitrate and ammonium are also important in controlling nitrogen flux in Loch Vale watershed in ice-free months. Similarly, virtually all hydrogen ion and nitrate inputs to the Isle Royale systems are retained in the terrestrial ecosystems.

Verifying Rates of Change in Surface Water Chemistry

The results of the NAPAP Long-Term Monitoring Project, initiated in 1982, have been analyzed. Data were used to estimate the number of lakes that should be sampled within a region to detect specified levels of chemical change at various levels of certainty. Analysis of variability demonstrated that among-lake variability exceeded annual and seasonal variability, indicating that sampling more lakes in a region would more accurately characterize regional trends than would sampling fewer lakes more intensively. These findings also increase confidence in the regional-scale index patterns developed in the NSWS. No changes have been detected, but the results are being re-analyzed in light of new information. These data are essential to the proposed Temporally Integrated Monitoring of Ecosystems (TIME) project, scheduled for implementation in 1990.

Biological Effects

Trends were evaluated in the water chemistry and angler harvest of Atlantic salmon in the only streams in the United States that support self-sustaining populations of the species. Results showed that Atlantic salmon resources in the northeastern United States appear to be presently unaffected by acidic deposition.

Burdens of certain potentially toxic metals (cadmium, lead, and aluminum) in brook trout were correlated with lake chemistry variables that are related to acidity. This study documents the occurrence of elevated trace metal concentrations in fish from low-pH lakes in Maine and New Hampshire, and shows a correlation between certain acidity-related water chemistry variables and whole-body metal burdens in fish. In a separate study (Little Rock Lake, Wisconsin), mercury concentrations in whole-body tissues of treatment-basin yellow perch were significantly elevated, and the growth rate of young-of-the-year largemouth bass was reduced.

Laboratory studies are continuing to define how acidity and such chemical constituents as calcium and aluminum affect fish. Studies on brook trout show that an increase in pH from 4.5 to 5.5 without an increase in calcium decreases fry mortality, but slow growth and impaired behavior patterns remain. Increases in pH beyond 7.0 lead to reductions in growth of brook trout. Other studies show that aluminum concentrations greater than 74 $\mu g\ L^{-1}$ cause gill abnormalities in salmon fry. Aluminum and pH also interact to affect the anatomy and activity of young smallmouth bass.

Experiments were also conducted that suggest the sensitivity of invertebrates to increased acidity. The acidification of experimental wetlands showed that black ducks are more likely to gain weight and survive under higher-pH conditions (pH 6.8 vs. pH 5.0). Since invertebrates constitute a primary food source for black ducks, these responses may reflect invertebrate mortality at lower pH. In Little Rock Lake, shifts in the relative abundance of several invertebrate taxa were observed in the treatment basin in response to a pH decrease from 6.0 to 5.6.

Effects of increased acidity on aquatic plants (phytoplankton and filamentous algae) have also been studied in the laboratory and in Little Rock Lake. The laboratory study suggests that the species dominating the phytoplankton community in the spring, when short-term pH depressions might occur, are less susceptible to decreased pH than one dominating in the fall. A filamentous algal mat appeared over 12 percent of the littoral zone of the treatment basin of Little Rock Lake in each of the first two years of the experiment, and water transparency has increased.

Indirect Human Health Effects

A one-year evaluation of the relationship between acidic deposition and cistern drinking water supplies was conducted. The water quality characteristics, including the trace elements cadmium, copper, lead and zinc, in rainwater cistern supplies representing an area receiving acidic deposition (western Tennessee and Kentucky, where bulk deposition pH was 4.5) were compared to cistern water chemistry in an area not receiving acidic deposition (St. Maarten in the Netherlands Antilles, where bulk deposition pH was 6.54). Mean volume-weighted pH for bulk deposition was two pH units higher and sulfate was 50 percent lower in the control region. Although rainwater was neutralized upon contact with cistern masonry in both regions, statistically higher levels of 11 constituents, including cadmium and lead, were found for homes in the area receiving acidic deposition. The lead is suspected to have been leached from the plumbing systems. Levels in standing tapwater were higher than in running tapwater at both locations.

Ecological Effects of Mitigation

Ten lakes in the Adirondack Mountains region were limed and stocked to examine the variability in effectiveness of mitigative liming and fish population response. Preliminary results show ANC, pH, and calcium increased in all lakes immediately after liming, but subsequent ANC levels were regulated by thermal stratification and flushing rates rather than initial dose rates. Survival of brook trout, as shown by post-liming bioassays, was high in all lakes compared to pre-liming survival. So far, there are no consistent differences in survival or growth between selected (increased acid tolerance) and unselected strains of stocked brook trout. Growth and condition of stocked trout appeared to be sensitive indices of both physiological acid stress and possible changes in food availability.

Forecasting Rates of Change in Surface Water Chemistry

DDRP studies investigated the degree to which systems to the north and south of the limit of recent glaciation in the eastern United States are at sulfur input-output steady state, i.e., sulfur inputs are balanced by sulfur outputs. This was done by analyzing available data on wet and dry deposition and comparing it to lake outputs, as calculated from data on runoff and lake chemistry. Watersheds to the north of the limit (approximately 42° N. latitude) appear on average to be at steady-state with respect to retention of atmospherically deposited sulfur. Systems not currently at sulfur steady-state will likely lose some ANC as they approach steady-state. The relationship between sulfur deposition, sulfur retention, and surface water chemistry is also being investigated. Some systems might respond by losing ANC through depletion of exchangeable base cations. The number of systems that will or will not respond is still being evaluated.

In contrast, watersheds to the south of the limit of glaciation appear, on average, to be retaining most of the sulfur deposited from the atmosphere. Changes in ANC associated with sulfur retention in the watersheds are also being investigated.

MAJOR DELIVERABLES

Deliverable	Date	Significance
Evaluation of variability in the chemistry of low-ANC lakes in the Northeast.	1988	Quantifies the spatial and temporal variability associated with the National Lake Survey - Phase I population estimates, based on the single fall index sample, and allows the Phase I estimates to be refined.
Evaluation of current status of of streams in the Middle Atlantic and Southeast.	1988	Provides regional population estimates of current chemical status of potentially sensitive stream reaches in two major regions of the eastern United States.
Estimates of the number of small (1-4 ha) acidic lakes in the Adirondack Park and an evaluation of differences in chemistry between large and small lakes.	1989	Provides refined estimates of the lake resource at risk as a result of acidic deposition in a subregion sampled during the Eastern Lake Survey - Phase I.
Analysis of evidence for past acidification of high-elevation lakes on glaciated sandy soils in Maine, and implications for future changes in ANC.	1989	Provides a means to estimate historical changes which may be attributable to acidic deposition and to evaluate the potential for future change in a subset of Maine lakes.
Book examining the current status, evidence of past changes, and the processes controlling surface water chemistry in selected regions of North America.	1989	Provides a comprehensive analysis of surface water chemistry and its relationship to acidic deposition using the National Surface Water Survey and other data bases for 12 regions in North America.
Report on response characteristics of lake and stream watersheds to sulfur deposition.	1989	Describes the potential long-term effects of sulfur deposition on the alkalinity of lakes and streams in the Northeast and Southern Blue Ridge Province.

Deliverable	Date	Significance
Journal article on the magnitude, duration, frequency, and potential importance of acidic episodes in streams of the Southern Blue Ridge Province.	1990	Provides detailed evaluation of the importance of short-term acidification from storm events in the Southern Blue Ridge Province.
Final report on sensitivity and uncertainty analysis for the Direct/Delayed Response Project (DDRP) models.	1988	Provides evaluation of the characteristics and behavior of the three DDRP models being used to forecast future acidification trends.
Estimation of the regional importance of hydrology, soil base cation supply, and other factors in acidification of surface waters in the Southeast.	1989	Provides data to assess the accuracy of predicted responses to acidic deposition for southeastern surface waters.
Projections for the recovery of the population of currently acidified surface waters in the Northeast and Southeast, based on a reduced level of sulfate deposition.	1989	Provides regional, quantitative estimates of the number of systems forecast to recover, given different levels of reduction in sulfate deposition.
Models for prediction of effects of effects of chronic acidification on fish populations in lakes in the Adirondack region of New York and the upper Midwest.	1989	Provides regionally applicable models for chronic effects of acidic deposition on fish populations, using chemical data from Adirondack and upper Midwestern lakes.
Journal article on the ionic dynamics of long-term snowpack and streamwater chemistry.	1988	Suggests factors that control which snowpack solutes actually enter streams.
Journal article on sulfate loss from small watersheds in the Upper Midwest.	1988	Investigates sulfate inputs, soil sulfate adsorption capacity, and surface stream water chemistry across a gradient of sulfate inputs.
Journal article on the importance of soil macropores in modifying snowpack melt chemistry	1988	Evaluates the importance of a potentially major factor determining the quantity of determining the quantity of snowpack ions entering streams.
Journal articles on soil properties of Loch Vale Watershed.	1988	Describes soils, chemical equilibrium relationships, and potential soil response to acidification.
Journal article on ionic inputs and outputs in a mixed conifer forest in the Sierra Nevada.	1988	Describes how ions associated with acidic deposition (H^+, NO_3, SO_4) accumulate while major nutrients (Ca, Mg, K, Na) are lost.

Deliverable	Date	Significance
Publication on weathering rates and soil kinetics in the Emerald Lake watershed of Sequoia National Park.	1988	Provides weathering rate equations for different subalpine soil types in a high-elevation, granitic watershed.
Report on artificial acidification of lake mesocosms and potential effects of acidic deposition on the community structure of zooplankton.	1988	Documents significant changes in the community structure of zooplankton resulting from slight changes in water chemistry due to acidic additions.
Report on the variation of wet and bulk precipitation chemistry along an elevational gradient in Sequoia National Park.	1988	Summarizes precipitation chemistry from stations at elevations of 518 m, 1767 m, and 2804 m.
A critical review of the uptake of mercury, cadmium, and lead by fish in low-pH waters.	1988	Discusses factors that cause elevated body burdens of these metals in fish in low-pH waters. Considers potential effects of increased trophic transfer of mercury, cadmium, and lead to birds and mammals.
Report on the acid sensitivity of cutthroat trout.	1989	Describes the sensitivity of cutthroat trout to low pH and elevated concentrations of aluminum.
Report on results of intensive studies of stream fish populations in Maine.	1988	Describes precipitation volume and chemistry, stream discharge and chemistry, and the species composition, production, and mortality of fish populations in six small streams in Maine.
Journal article on the use of benthic macroinvertebrates as indicators of the sensitivity of stream communities to acidification.	1989	Describes a technique that uses the combined presence of certain macroinvertebrate taxa to to evaluate stream communities in the Southern Blue Ridge Province for sensitivity to acidification.
Journal article on bone development in fish in Maine lakes with varying levels of acidity.	1989	Describes the effect of low pH conditions on bone strength and bone development in fish.
Journal article describing a cadmium-saturation technique for measuring metallothionein in fish.	1989	Presents a method for quantifying various protein fractions associated with the metal-binding protein metallothionein in fish, an approach used to assess stress from certain metals mobilized by acidic deposition.
Journal article on metallothionein as an indicator of cadmium-induced stress in brook trout.	1988	Evaluates the measurement of metallothionein in fish and discusses the utility of the method for assessing cadmium-induced stress in a species that is widespread in low-ANC waters of the northeastern United States.

Deliverable	Date	Significance
Journal article on the influence of low pH and aluminum exposure on the early life stages of Atlantic salmon.	1988	Describes the effects of low pH alone and with various concentrations of aluminum on early life stages of Atlantic salmon.
Journal article on chronic, no-observed-effect concentrations of aluminum for brook trout exposed in dilute acidic water.	1988	Establishes chronic, no-observed-effect concentrations of aluminum for brook trout exposed to the metal at pH 5.5 and 6.5.
Final report on effects of acidification on free-living wetland wildlife.	1988	Species abundance and diversity as well as specific habitat use is assessed for waterfowl and water birds in regions of low and high acid neutralizing capacity bedrock.
Final report on ecosystem effects of experimentally acidified wetlands.	1988	Assesses the potential impacts of lower pH and possible mobilization of toxic metals in wetlands on growth of black duck ducklings.
Final report on the effect of cations mobilized by acidic precipitation on the productivity and survival of terrestrial vertebrates.	1989	Provides information on the food chain response to mobilized cations in acidified environments.
Final report on declining amphibian populations in the Colorado Front Range.	1988	Provides baseline information on the potential sensitivity of certain western amphibians to acidic deposition and information on status of amphibian populations in potentially sensitive areas in the Rocky Mountains.
Acid Precipitation Mitigation Guidance Manual.	1988	Guidance manual for program conducting mitigation research on lakes and streams. Includes standardized protocols for physical, chemical, and biological sampling.
Final report on remedial action to reverse fish reproductive failure in response to acidic conditions in surface water.	1988	Provides information on the most sensitive life stage of brook trout to acid/metal interactions. Discusses the effectiveness of limestone emplacements for increasing tolerance of brook trout in the early life stages.
Fishery management guidelines for mitigating (liming) acidified surface waters in the Adirondack region.	1988	Provides information to assist managers in the restoration of fisheries in the Adirondack region.
Final report and model on thermal stratification effects of limed lakes.	1988	Models the effects of base addition on light attenuation and thermal stratification, both of which have a strong influence on lake biology.
Journal article on relationship between acidic deposition and and cistern drinking water supplies.	1988	Describes potential effects of acidic deposition on cistern and drinking water supplies and human health by comparing deposition and cistern water quality in an area receiving acidic deposition to an area where deposition is not acidic.

EFFECTS ON MATERIALS AND CULTURAL RESOURCES

INTRODUCTION

Air pollution has been suspected since the mid-19th century of accelerating the degradation of many types of construction materials and cultural artifacts. Various air pollutants contribute to this increased damage through wet and dry acidic deposition. The extent of this damage is unknown, and to provide a basis for informed decisionmaking, several factors need to be better defined: the amount and rate of materials damage attributable to wet and dry acidic deposition above the natural rate of degradation, a quantitative evaluation of the processes involved, and a reliable estimate of the quantities of materials at risk. Ultimately, it will be necessary to understand how the increase in the rate of deterioration caused by pollutants reduces the service life and economic value of materials.

Our knowledge of the deterioration of man-made structures and building materials caused by acidic deposition in both urban and remote sites is limited both by a lack of previous research and by our ability to relate observations of materials deterioration to environmental data from pollution monitoring. While deposition-related effects on building materials, structures, and works of art have been reported, neither the dose-response relationships, the overall extent of those effects, or their economic implications have been established. Quantifying the effects of acidic deposition on man-made structures is further complicated by the difficulty in estimating economic impacts resulting from the predicted rates of acidic damage.

1990 RESEARCH OBJECTIVES

- Quantify the incremental effects of acidic deposition on material service lives relative to material degradation under natural background conditions (dose-response functions).

- Estimate the quantity and geographic distribution of materials that are significantly impacted by acidic deposition above natural background levels.

- Estimate the economic impacts of reductions, where measurable, in material service lives due to acidic deposition.

- Evaluate the feasability of substitution, protection, and other strategies to mitigate materials damage.

RESEARCH ACTIVITIES IN 1987

In 1987 the Task Group's highest priority has been to focus on the development of dose-response functions. Effort toward meeting the other objectives must increase as dose-response research is completed.

Field Test Sites

The five field test sites in the northeastern United States continued operation in 1987. Research at these sites (Washington, DC; Research Triangle Park, North Carolina; Chester, New Jersey; Newcomb, New York; and Steubenville, Ohio) provides for detailed measurements of environmental exposure (in cluding meteorological data as well as air and rain concentration data of relevant acids and associated compounds) and of the degradation of galvanized steel and other metals, carbonate stone, and paint systems. Movable covering devices at two of the sites are used to examine the effects of wet and dry deposition, an important step in the development of dose-response functions. In addition to exposure under ambient environmental conditions, the sheltering facilities at the Research Triangle Park site have been enhanced by the installation and operation of acid rain simulators which allow the manipulation of simulated rainfall acidity in contrast to ambient conditions. Further, the orientation of metal and paint samples at the field sites is being examined to determine how varying exposure (vertical, horizontal, north- or south-facing, etc.) affects degradation of these materials. These measurements provide a basis to translate observations about materials degradation based on highly controlled laboratory experiments to observations in the ambient environment.

Laboratory Research

The mechanisms of wet and dry deposition in the corrosion of metals, dissolution of carbonate stone, and deterioration of paint systems are being studied and quantified in controlled chamber experiments. The chambers can control levels of sulfur dioxide, rain acidity, surface moisture, temperature, and relative humidity. Dose-response functions for rain acidity, sulfur dioxide, and nitrogen oxides concentrations are being derived from these controlled laboratory experiments. Laboratory results are being compared to long-term exposure experiments at the field sites to provide a firm basis for damage function development.

Research on paint systems examines both latex and oil-based paints on wood and metal substrates. The work emphasizes early microscopic predictors of subsequent macroscopic paint failure.

Special Studies

Special studies focus on measuring the distribution and deterioration of cultural and construction materials in current use. These include determination of the variation in microclimate and deposition around building surfaces and how dose-response functions can be applied to complex structures. These studies will provide the refinements necessary for the appropriate application of dose-response functions developed in the laboratory and modified by field test results.

Inventory, replacement patterns, and replacement costs of galvanized steel are being enumerated. If the results of these studies have acceptable confidence limits, estimates of the economic costs of damage due to acidic deposition may be made.

FUTURE RESEARCH ACTIVITY

Since the laboratory and manipulated field experiments on galvanized steel were largely completed in 1987, the emphasis within the metals program will shift to data interpretation. The principal focus within the stone research program will be on manipulated experiments at the field test sites, designed to differentiate the effects of wet and dry deposition and to quantify the effects of incremental levels of pH during wet deposition. Laboratory experiments initiated in 1986 are expected to provide a mechanistic understanding of carbonate stone dissolution for the development of dose-response functions. Deposition velocities of atmospheric precursor gases on carbonate stone surfaces will be determined using wind tunnel experiments. Experimental work on paint/substrate systems will continue in 1988. Laboratory and field experiments will focus on deposition processes, the solubility and penetration of pollutant species through polymer coatings, and chemical changes at the paint/substrate interface. Planning will begin in 1988 for work in 1989 and 1990 to apply the results of the research on dose-response functions to the calculation of damage to typical structures.

1987 RESEARCH HIGHLIGHTS

- Field data on the atmospheric corrosion of zinc and galvanized steel (steel coated with a protective layer of zinc) indicate that the corrosion process involves the formation of a two-layer corrosion product with: (1) an inner layer sensitive to gaseous pollutants (particularly sulfur dioxide) affecting the corrosion, and (2) a relatively non-protective outer layer that continues to grow even in long exposures and is affected by environmental factors such as wet deposition of hydrogen ion. Significant amounts of sulfur, as much as 50 percent of the total deposition, are delivered to the zinc and galvanized steel surfaces by dry deposition. The corrosion of zinc and galvanized steel is characterized by rapid initial corrosion (first month), a decline in the corrosion rate during the transition to long-term behavior (one to three months), and then an increase in corrosion rate proportional to sulfur dioxide levels in long-term exposures (greater than one year). Loss of zinc from the corrosion film is proportional to the hydrogen ion load (rain hydrogen ion concentration times rain volume). Sheltering from rain leads to much lower corrosion rates (one-fourth to one-tenth of those for boldly exposed panels) despite the higher sulfur concentrations in the corrosion film which result from the absence of surface washing under the shelter. The lower time of wetness under the shelters is one of the major causes of the reduced corrosion rates. Other results from zinc panels suggest that electrochemical coupling between the skyward and groundward sides of panels produces an overall reduction in the corrosion rate.

- In addition to the field site experiments described in the previous section on zinc and galvanized steel, laboratory chamber experiments have been used to study the dissolution of zinc corrosion product as the result of both the wet deposition of rain of varying acidity as well as the dry deposition of sulfur dioxide. The experiments have provided the following results:
  - During periods of surface wetness, sulfur dioxide reaching the surface reacts in a one-to-one relationship with the zinc corrosion film.
  - Hydrogen ions react in a two-to-one ratio with the zinc corrosion film.
  - The corrosion film of basic zinc carbonate is soluble in clean rain. The dissolution depends on the residence time of rain on the surface.
  - During periods of surface dryness sulfur dioxide will saturate on the surface until a single molecular layer of sulfur dioxide is formed.

An automatic covering device, installed at the test site located at the Research Triangle Park, North Carolina, is being used to enhance the development of dose-response relations that partition the effects of wet and dry deposition on materials. The apparatus provides for exposures of two sets of test panels under separate conditions: ambient rain, and automatic spraying with de-ionized water to simulate rain with a pH of 5.6. Chemical analysis of the run-off from the test panels on a rain event basis provides an immediate measurement of zinc dissolution and the deposition of sulfur oxide and nitrogen oxide species to the extent that they are soluble and are collected in the run-off. Analysis of the ambient rain run-off solutions provides a measure of zinc loss due to dry and wet deposition, while analysis of the deionized spray run-off provides a measure of zinc loss due to sulfate deposited by dry deposition and zinc loss due to the solubility of the corrosion layer in "clean rain" (rain saturated with carbon dioxide). An evaluation of the run-off data from the automatic spray/covering device at the North Carolina exposure site has shown that controlled field studies can be used to partition the contributing factors that influence the dissolution of zinc corrosion products. The preliminary results from these experiments suggest that less than 30 percent of the loss of zinc carbonate corrosion product from galvanized steel surfaces is due to dry deposition, less than 20 percent is due to wet deposition, and more than 50 percent is due to the solubility of the corrosion layer in "clean rain" under the environmental conditions at Research Triangle Park.

A theoretical model has been developed for determining the cumulative atmospheric corrosion of galvanized steel based on the results from laboratory and controlled field studies described above. The model takes into account the growth and dissolution of the corrosion product layer in the presence of dry deposition of sulfur dioxide, acidic deposition, and clean rain. The current model does not take into account the potential neutralizing capability of dry deposited alkaline particles or the impact due to the dry deposition of other acidic gases such as nitric or organic acids. The model is being testedusing the weight loss data from the field exposure experiments coupled with the data available in the aerometric data base from the field test sites. The experimental work is being extended to include analyses of the run-off from panels sprayed with solutions of varying pH in order to test the incremental loss of corrosion product due to hydrogen ion concentration. Run-off collections are also being analyzed from galvanized steel materials of different shapes and orientations. These experiments should provide insight into the effects of acidic deposition to galvanized steel structures.

- Using acid concentrations down to pH 2, which are even more acidic than those found in highly acidic fogs, NAPAP researchers have shown that brief exposure of wood to acid during experimentally accelerated weathering increased the erosion rate. The erosion rate was determined by weight loss and was affected by the strength and type of acid. Under the most severe acidic condition, the wood surface eroded twice as fast as a similar surface that was exposed to distilled water. This accelerated weathering will shorten the service life of unpainted wood and may also affect the performance of painted wood. The adhesion of paint to wood is strongly affected by the amount of weathering the wood receives prior to painting. In the absence of acid precipitation, wood weathered more than four weeks prior to painting had decreased paint adhesion. Weathering under acid conditions may further decrease the time wood can be left exposed between surface preparation and painting, without sacrificing paint performance.

A series of short-term exposure experiments in a laboratory test chamber was conducted to study the deposition of acidic gases on painted surfaces. Preliminary analyses of the data suggest that the uptake of sulfur dioxide into thin layers of moisture is controlled by the amount of alkaline materials in the coating (i.e., the buffering capacity of the paint) and that the uptake into dew-covered painted surfaces is substantially below that of damp galvanized steel. The uptake of nitric acid is less sensitive to the alkaline nature of the surface coating than sulfur dioxide. Similar observations have been made in laboratory experiments on carbon steel panels covered with oil-based paint, and on latex-painted wood panels.

- One of the primary modes of environmental damage to carbonate stone buildings is the formation of gypsum deposits, particularly on sheltered surfaces, due to the reaction of atmospheric sulfur oxides with calcium in the stone. Gypsum has also been observed to form on the sheltered undersides of the test briquettes exposed for one or two years at the NAPAP materials exposure sites. The presence of gypsum on the downward exposed surface of the samples has been confirmed by chemical analysis, X-ray analysis, near infrared spectroscopy, and scanning electron microscopy.

Surface chemistry results for the first few years of exposure show that gypsum accumulation is nearly linear with time. The highest average annual sulfate accumulation observed on marble test briquettes was approximately 200 micrograms/square centimeter, and was roughly correlated with ambient sulfur dioxide levels at the test sites. Sulfate accumulation on limestone was 2-5 times the amount gained by marble at all sites due to the greater porosity of the limestone.

- Laboratory research during 1987 focused on the simple air/water/carbonate stone system, which must be studied in order to understand the more complex wet/dry and added sulfur dioxide systems. In the simplest system, three independent processes operate, any one of which can be slow enough to control the rate of weathering and therefore warrant inclusion in the dose-response function: 1) the exchange rate of carbon dioxide gas between the air and the wetted surface of the stone, 2) the rate of conversion of dissolved carbon dioxide to hydrogen ion and bicarbonate and the reverse reaction, and 3) the rate of dissolution and precipitation of calcium carbonate in the stone. A computer model of these processes was developed using published rate constants for the reactions involved, and the model was confirmed by experiment.

Two different experimental approaches were taken to isolate and separate the weathering of carbonate stone due to dissolution at the surface and internal degradation resulting from dissolution within stone pores. Surface rates of dissolution of limestone and marble were measured and compared to rates predicted for pure calcite based on previously published experiments. It was concluded that the experimentally determined surface dissolution rates were consistent with predicted rates based on theoretical models, and that the models could adequately predict the solution rate of carbonate surfaces by acid rain standing on those surfaces. Current experiments and modeling are examining the solution rate by a moving film of water.

The potential for internal degradation resulting from dissolution in stone pores was investigated by forcing solutions through cylinders of Salem limestone at pressures up to three bars, at flow velocities up to about 0.01 cm/sec. Chemical changes in the pore water were measured within the apparatus using a specially designed specific conductance electrode and a pH electrode. The rapid saturation of internal solutions with calcium (less than 13 seconds) and the rapid movement of water into the stone interstices suggest that only minor internal damage to the limestone and marble can be expected to result from their chemical attack by sulfur-dioxide-free rain.

- Erosion (or mass loss) of samples of Shelburne (Vermont) marble and Salem (Indiana) limestone exposed to ambient environmental conditions at four materials exposure sites since 1984 has been measured by alternative methods, including: (1) analysis of excess calcium in the rain run-off from samples compared with analyses of incident rain, (2) spectroscopic analysis of surface recession and roughness, and (3) monitoring the changes in stone specimen weights. Results from the first year of measurements by these three methods yielded similar results for marble. Runoff data from the North Carolina site yielded a value of 14 micrometers of stone loss per year; direct spectroscopic measurements of surface recession yielded a value of 13 micrometers per year; and weight loss measurements provided an estimate of 16 micrometers per year if the loss is assigned entirely to the skyward surface of the specimens. Recession values of this magnitude were also measured for the New Jersey, New York, and Washington, DC sites, but with somewhat larger uncertainties. Limestone weight loss was anomalously higher than that of marble during the first year of exposure, possibly as a result of the loss of powder residues and extraneous material entrained in pores after sample fabrication, but limestone losses approached the magnitude of those of marble during the second year of exposure. Samples prepared from weathered limestone and marble removed from a building 20 to 80 years old, respectively, have been prepared for addition to the field exposure sites in order to examine their response to deposited pollutants relative to the freshly quarried stone specimens.

- In-situ monitoring of the effects of atmospheric pollutants to materials of cultural significance are under way in four separate experimental programs:

1. A comparative analysis of the corrosion of a series of bronze statues cast from the same model in the early 20th century and placed in various locations across the country is in progress. Over fifty copies of *The Hiker* statue were erected in the United States between 1900 and 1965, primarily in the 1920's and 30's. Detailed documentation of the extent of corrosion has been completed, and corrosion samples collected for analysis. An attempt will be made to correlate the corrosion data with estimates of historical pollutant dose and local environmental factors.

2. A direct analysis of material removed from bronze and marble monuments by rainfall is being conducted at Gettysburg National Military Park, Pennsylvania. Four bronze and four carbonate stone monuments are currently being studied. Runoff from each of the monuments is collected by the Park staff for each rain event and analyzed chemically. The analytical data are being related to meteorological and gas/particle concentration data collected at two locations in the park.

3. Test walls of sandstone masonry blocks which were once part of collapsed Anasazi ruins at Mesa Verde National Park are being used to determine if increased pollution may accelerate the weathering of the nearby cliff dwelling ruins. Photomicrographs are made of reference areas on each block in the test walls at regular intervals and used to define recession rates which can then be related to environmental factors measured on site.

4. An early 19th-century marble building at Independence National Historical Park, Philadelphia, is being used as a prototype for studies related to the variations in the weathering of stone buildings due to orientation and sheltering. Time of wetness is being monitored continuously at various locations on the building in order to relate this important parameter to variations in exposure and the degree of sheltering. Monitoring of particle and gas concentrations at selected points on the building (using filter packs triggered by the presence of surface moisture) will begin in February 1988. Stereophotogrammetry has been used to document the current condition of the local and Carrara marbles used in the structure and to estimate the variation in marble loss as a function of exposure.

MAJOR DELIVERABLES

Deliverable	Date	Significance
Report on dose-response functions on selected metals	1988	Presents first-generation dose-response functions produced under laboratory conditions for the effects of pH, sulfur dioxide, and oxides of nitrogen on selected metals.
Report on distribution of surface coatings	1988	Presents a distribution and use of surface coatings by coating type and region. Defines economically important paint/substrate systems for study, and the important macroscopic modes of failure of exterior finished surfaces.
Report on distribution of galvanized steel	1988	Presents a distribution and average service life of galvanized steel by application and region.
Report on analytical techniques for measuring acid deposition effects on a variety of coated surfaces	1988	Describes analytical techniques to measure specific degradation processes of polymer coatings including the rate of penetration of acidic species to substrate, rates of autooxidation, cross linking and chain scission in the film former, rate of embrittlement of the film former, and the failure strength of paint film.
Report on the application of damage functions for zinc and galvanized steel to field exposure data: integrative approach	1988	Presents an analysis of the data from the long-term exposure (greater than one year) of zinc and galvanized steel test panels at the field exposure sites. Integrates the corrosion data base with rain chemistry and aerometric data for each site.
Report on the application of damage functions for zinc and galvanized steel to field exposure data: differential approach	1988	Presents an analysis of the data from short-term exposure (less than one year) of zinc and galvanized steel test panels at the field exposure sites. Defines the chemical processes occurring at metals surfaces with short-term variations in environmental parameters.
Report on statistical analysis and interpretation of aerometric data for the field exposure sites	1988	Presents the environmental data from the five materials exposure sites that have been assembled, stored, and validated in a uniform aerometric data base. The data include air quality (SO_2, NO_x, O_3) and meteorological parameters (wind speed, wind direction, relative humidity, temperature, precipitation, and solar radiation).

Deliverable	Date	Significance
Report on acid deposition to coated surfaces	1988	Presents the results of laboratory chamber studies of the effects of acid precursor gases and acid deposition on selected paint/substrate systems.
Report on the effects of deposition on paint/substrate systems	1989	Presents the initial results from field exposure experiments on the micro/macro effects of acid deposition on a variety of paint/substrate systems.
Final Plan for Historic District Field Survey: northeastern states	1989	Describes a plan for field validation of the distribution of historic stone buildings in the northeastern United States.
Report on laboratory studies relating to carbonate dissolution processes	1989	Presents the interpretation of kinetic experiment results, atmospheric reaction chamber results, and a geochemical model for pollutant-related dissolution of carbonate stone.
Report on laboratory studies relating to the deposition of atmospheric precursor gases to stone surfaces	1989	Describes the deposition rates of SO_2 and NO_x to the surfaces of various fresh and weathered carbonate stone under ambient environmental conditions.
Report on dose-response functions for selected coating systems	1990	Presents first-generation dose-response functions produced under laboratory and field conditions for the effects of pH, sulfur dioxide, and oxides of nitrogen on polymeric surface coatings.
Report on dose-response functions for carbonate stone	1990	Presents the algorithms to be used for the dose-response functions for carbonate building stone (including information on changes in surface composition, roughness, recession, and run-off chemistry) in an economic assessment of effects of acidic deposition.
Report on the distribution of historic buildings in the Northeast	1990	Presents a data base of the distribution of historic stone architecture in the Northeast, based on the index of stone uses and field verification surveys.
Report on the distribution of monuments and outdoor sculpture east of the Mississippi River	1990	Presents a data base of the distribution of monuments and outdoor sculpture east of the Mississippi River, based on American Monuments and Outdoor Sculpture (AMOS) and field verification surveys.
Final report on *The Hiker* project	1988	Describes the relative degradation of replicate bronze statues exposed for 40 to 60 years to various environments.

Deliverable	Date	Significance
Report on Mesa Verde Study	1989	Describes the results of four years of parallel monitoring of sandstone decay and environmental factors.
Report on Gettysburg Study	1989	Describes the results of in-situ run-off experiments on bronze and carbonate stone monuments at a rural Pennsylvania site.
Report on Independence Study	1990	Describes the results of two years of parallel monitoring of marble decay and environmental factors at an urban Philadelphia site.
Reports on 5-year field exposure tests of carbonate stone	1990	Describes the results of mineralogical, surface chemistry, roughness/recession, weight loss, color change and run-off measurement at five field sites in relation to environmental data supplied by the EPA.

PUBLICATIONS

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